UNIVERSITY OF BIRMINGHAM

CHARACTERISATION OF PARTICULATE MATTER EMISSIONS FROM COOKING

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Abstract

Cooking fume have been found to be a significant component of ambient particulate matter and also to contribute to high concentrations of aerosol indoors. As several studies have linked exposure of individuals to cooking emissions with adverse health effects, the need to further understand the composition of this source of particulate matter is essential. It has also been identified that there is a gap in literature for existing cooking profiles in the UK with the few existing ones being generated a long period ago in different geographical locations, with none obtained locally to the UK.

This study was concerned with gaining further insights into the chemical composition of aerosol generated from typical styles of cooking and the understanding of trends of the formation of particles among different culinary methods. Cooking source profile for African, Chinese, Western and Indian styles was obtained in a specially designed laboratory based kitchen. These profiles were used as input in a Chemical Mass Balance model where ambient data collected in Birmingham, UK were analysed in order to apportion the quantity of organic matter from cooking sources in the location sampled.

Measurements of particulate matter were also collected while cooking in a real kitchen in order to quantify magnitude of particulate matter emissions in a household setting generated from cooking.

It was found that cooking generated a significant mass of aerosol with the particle sizes largely within the respirable size range. Also the major groups of chemical compounds which were identified from cooking included alkanes, acids, Polycyclic Aromatic Hydrocarbons, glycerides and sterols. The Chinese style of cooking was found to generate the highest concentration of particles with PM mass of 21.61μg/m^3. African style cooking had the lowest

total concentration when compared to all other styles. The main group of compounds released during Indian and western style alkanes with PAH having the highest concentration when the total emitted compounds for African cooking was analysed (6.83 µg/m^3). High concentrations of monoglycerides were observed in Western and Chinese cooking, 10.33µg/$m^3$and 11.52µg/m^3 respectively.

Stir frying and grilling were found to generate the highest particulate matter concentration compared with stewing. Cooking involving the use of meat was found to generate more particles. The profiles of compounds emitted in the real kitchen were similar to those of the trailer.

The source profile from cooking (a range of marker compounds attributed to cooking) obtained from the study were found to correlate well with each other with Indian and Western profiles exhibiting the highest correlation. When used for the CMB model runs, these two profiles provided the best output with the model runs apportioning 16% of the Organic Carbon to be from cooking, with traffic, wood smoke and soil debris contributing 44%, 18% and 24% respectively.

The cooking experiments involved only rice chicken and potatoes, future work can include additional ingredients and other cooking styles.

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Abbreviations

AQEG	Air Quality Expert Group
BSTFA-TMCS	Bis-trifluoroacetamide trimethylchlorosilane
CMB	Chemical Mass Balance
COD	Coefficient of Divergence
DCM	Dichloromethane
DEFRA	Department for Environment, Food and Rural Affairs
DR	Diagnostic Ratio
EC	Elemental Carbon
EUSAAR	European Supersites for Atmospheric Aerosol Research
FID	Flame Ionization Detector
GBD	Global Burden of Disease
GC-MS	Gas Chromatography-Mass Spectrometry
IARC	International Agency for Research on Cancer
LoD	Limit of Detection
LPM	Liters per Minute
MPIN	Modifed Pseudo-Inverse Normalized Matrix
NAAQS	National Ambient Air Quality Standards
NAEI	National Atmospheric Emissions Inventory
NIST	National Institute of Standards & Technology
OC	Organic Carbon
OM	Organic Matter
PAH	Polycyclic Aromatic Hydrocarbon
PCA	Principal Component Analysis
PFTBA	Perfluorotributylamine
PM	Particulate Matter
$PM_{2.5}$	Particulate matter with aerodynamic diameter less than 2.5 µm
PM_{10}	Particulate matter with aerodynamic diameter less than 10 µm

PMF	Positive Matrix Factorization
POC	Primary Organic Carbon
PTFE	Polytetrafluoroethylene
RM	Receptor Model
SOC	Secondary Organic Carbon
SPM	Suspended Particulate Matter
SPSS	Statistical Package for Social Sciences
TMS-DM	Trimethylsilyldiazomethane
TC	Total Carbon
UFP	Ultrafine Particle
UoB	University of Birmingham
USEPA	United States Environment Protection Agency
VOC	Volatile Organic Compound
WHO	World Health Organization

Chemical mass balance code.

CODE	SPECIE NAME
BZKFLU	benzo(k)fluoranthene
BZEPYR	benzo(e)pyrene
BZAPYR	benzo(a)pyrene
PYR	perylene
INDFLU	indeno(123cd)fluoranthene
INDPYR	indeno(123cd)pyrene
DBZANT	dibenz(ah)anthracene
PICENE	picene
BZGHPL	benzo(ghi)perylene
CORON	coronene
PALMTA	Palmitic acid (n-hexadecanoic acid) (C16)
LINOLA	Linoleic acid (9,12-Octadecadienoic acid)
OLA	Oleic acid (9-Octadecenoic acid) (C18:1)
STEARA	stearic acid (n-Octadecanoic acid) (C18)
LEVOG	levoglucosan
CHOL	cholesterol
UNDEC	undecanoic
OCTA	octanedioic
DODE	dodecanoic
NONDIA	nonanedioic
TRI	tridecanoic
TETDE	tetradecanoic
PENT	pentadecanoic
HEP	heptadecanoic
NONA	nonadecanoic
EICO	eicosanoic
DOCO	docosanoic
TETCO	tetracosanoic
MONMY	1-Monomyristin
MONPA	1-Monopalmitin
MONOL	1-Monoolein
MONSTE	1-Monostearin

Declaration

I, Lami Karimatu Abdullahi, declare that this work was carried out completely by me. I carried out all the cooking exercises, filter sampling and chemical analysis (which included sample chemical extraction, GCMS analysis, OC/EC analysis) for the project. The XRF analysis of filters was out sourced and carried out in the Chemistry department of University of Birmingham.

CHAPTER 1 - Introduction and literature review.

This chapter gives a general introduction of particulate matter and cooking aerosols. The research aims and objectives are also discussed.

This chapter contains some sections of verbatim text adapted from the following review article published as part of this PhD:
Abdullahi, L, Delgado Saborit, JM & Harrison, RM 2013, 'Emissions and indoor concentrations of particulate matter and its specific chemical components from cooking: A review' **Atmospheric Environment**, vol 71, pp. 260- 294.

1.1 Particulate matter

Particulate matter (PM) is defined as the mass of a mixture of solid particles and liquid droplets of various sizes (range from a few nanometres to tens of micrometres) suspended in a volume of air which represent a broad class of chemically and physically diverse substances. Particulate matter is classified according to its size, thus PM_{10} is defined as the concentration of particulate matter with aerodynamic diameter of 10 micrometres or less, while $PM_{2.5}$ is defined as the concentration of particulate matter that has aerodynamic diameter of 2.5 micrometres or less. The average human hair is about 30 times larger than a large fine particle as hair is about 70 micrometers in diameter as illustrated in Figure 1(USEPA, 2013).

Particulate matter consists of components that are released directly from a source (primary PM) or are formed by chemical reactions in the atmosphere (secondary PM). It comes from natural and man-made sources and consists of a range of chemical compounds which can be useful for the identification of the source.

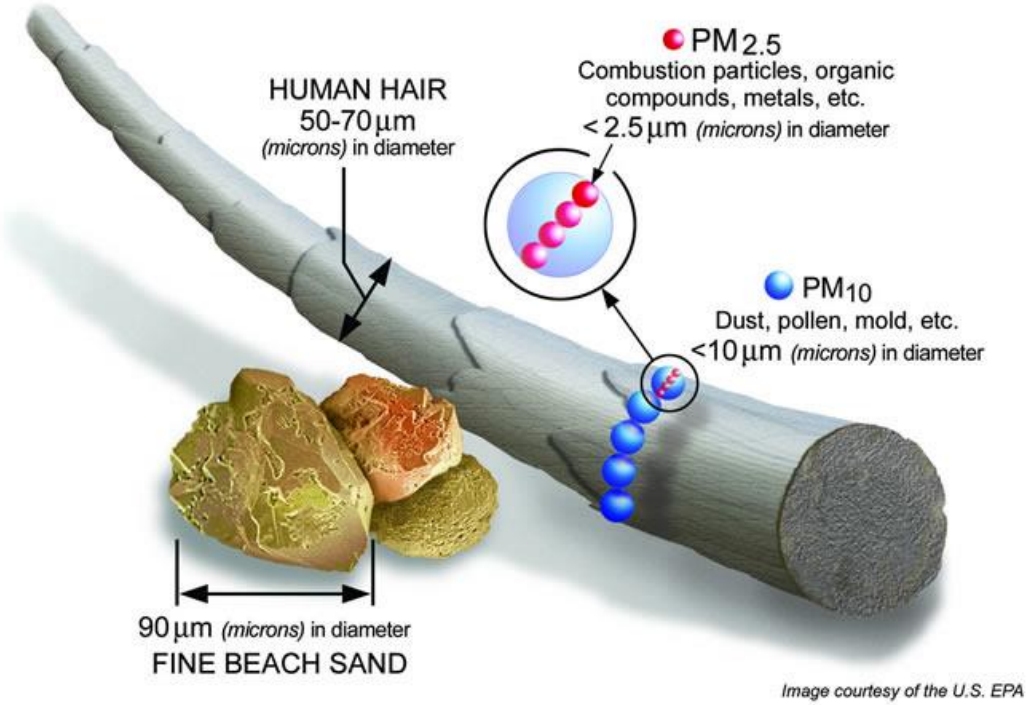


Figure 1 Particulate matter size (USEPA, 2015).

Primary PM is released from sources which include road transport (tyres and brake wear, engine combustion, road dust), industrial, commercial and domestic burning of fuels and also dust from these activities and natural sources (sea spray and dust). It generally consists of sodium chloride (from sea salt), trace metals (from metallurgical and mechanical abrasion processes - include lead, cadmium, nickel, chromium, zinc and manganese); elemental and organic carbon (from high temperature combustion of fuels and it consists of several individual components such as polycyclic aromatic hydrocarbons (PAH), alkenes, aldehydes) and mineral dust (coarse dust from quarrying construction and wind- include aluminium, silicon, iron and calcium).

Particle mass and number are generally the important metrics used to represent PM concentrations in exposure assessment (Harrison et al., 2010). Particle surface area is another metric of interest as it has been found that for the same mass of particles, the particle number

and surface area increase with decrease in particle size (Harrison et al., 2000). This has been observed in some toxicological studies such as Oberdorster, 2000, where it was found that particles become more toxic per unit mass as their size decreases.

Generally concentrations of PM are represented in terms of mass, number or volume, with mass being the most commonly used parameter. The use of mass is greatly for uniformity and comparability as the epidemiological studies from which the air quality standards are derived have used mass as the measure of particle concentration (Shi et al., 2001).

Particles are described using size distribution consisting typically of 4 modes which are:

- Nucleation (particle diameter less than 10 nm)
- Aitken (particle diameter between 10 and 100 nm)
- Accumulation (particle diameter between 100 nm and 2 μm)
- Coarse (particle diameter more than 2 μm).

For classification using the minimum in the mass size distribution particles are classed as:

- Fine particles consists of Nucleation, Aitken and accumulation mode;
- the Ultrafine particles consists of nucleation and Aitken modes and
- the Coarse particles consist of particles with diameters above 1 μm (Colbeck and Lazaridis, 2010).

Generally speaking and for regulatory purposes (using the fixed size cut off of 2.5 μm) particles with aerodynamic diameters less than 2.5μm are classified as fine particles and particles with aerodynamic diameters more than 2.5 μm are classified as coarse particles.

Fine and coarse particles generally differ due to their formation mechanisms, sources, chemical composition, and removal processes (Harrison et al, 2001). Fine particles are emitted during combustion processes or secondary aerosol formation while coarse particles are generated due to abrasion and mechanical processes (fugitive dusts from industrial sources, tyre-wear debris, re-suspended soils and street dusts, sea salts, pollen and fungi spores).

Processes such as condensation, evaporation and coagulation lead to changes in the size and composition of particulate matter in the atmosphere.

Condensation can occur with gaseous vapours whereby they combine with existing small nuclei (condensation nuclei) to form aerosols which can then grow in size by colliding and sticking together through the process known as coagulation. Particles can also be formed by nucleation which is the process where gases interact and combine with other molecules. Figure 2 offers a schematic representation of a typical size distribution for atmospheric particles, indicating some formation pathways.

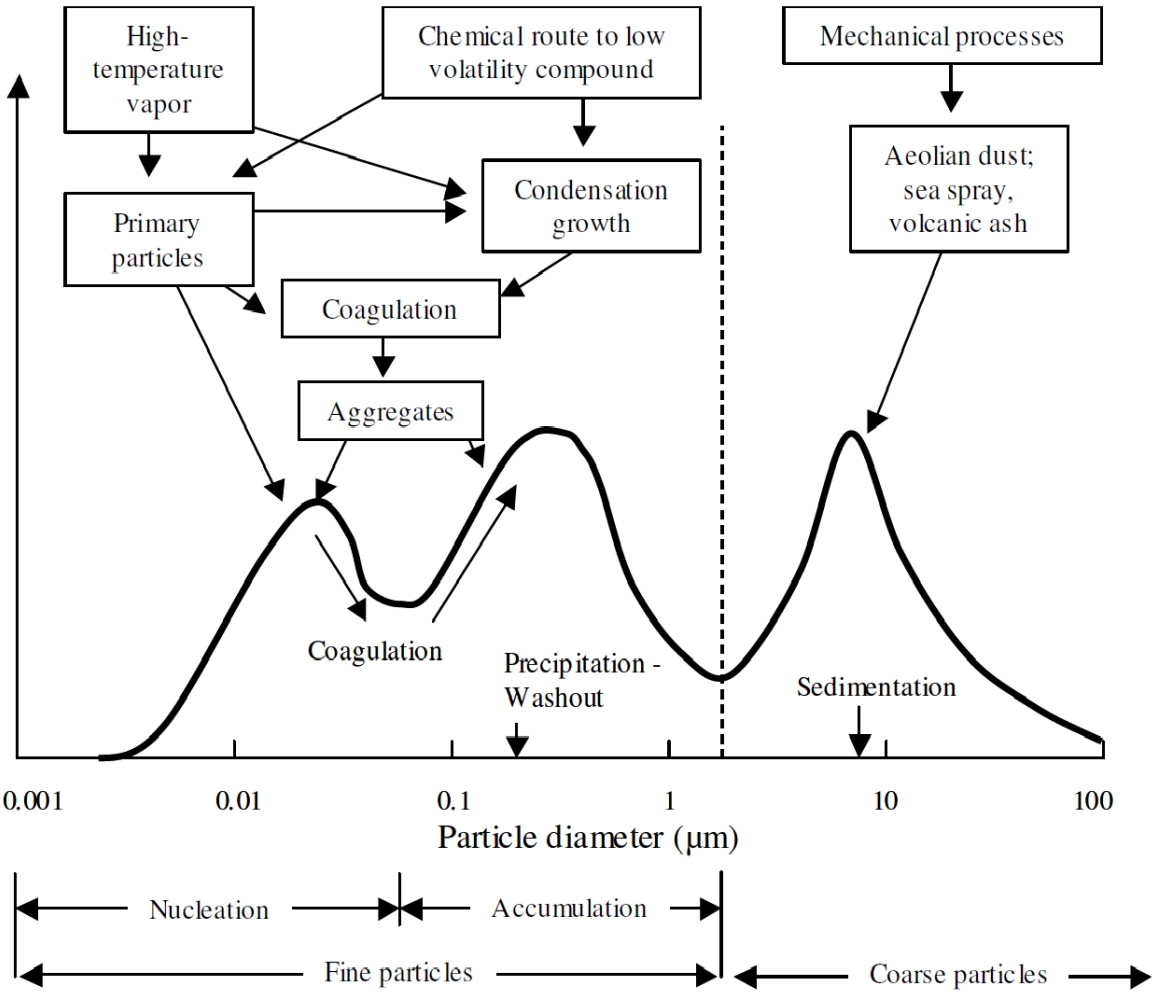


Figure 2 Schematic representation of a typical size distribution for atmospheric particles, indicating some formation pathways, (DEFRA, 2004).

The different fractions can exist in the atmosphere for varied periods ranging from minutes to weeks depending not only on the particle size but on other conditions such as the weather. Generally the particle size distributions can be used to identify particle sources which will help to understand the health implication of different source types which can aid in policy making and choices for abatement options.

For instance from monitoring data and various studies there has been a general decline of total emission of primary PM_{10} observed in the UK which has been mostly due to improvement in technology and change to cleaner fuel for general combustion in industries, domestic heating, energy production and road transport. Emissions inventory of PM_{10} from the UK have actually shown that domestic and commercial emissions have declined with emissions of PM_{10} falling from 263 kilotonnes (54% of total emission) in 1970 to 17.9 kilotonnes (20.9%) in 2012 (NAEI, 2014). Even with this observed decrease in concentrations, the UK residential sources have still been found to represent a high percentage (13%) of the national primary, man-made emissions of $PM_{2.5}$ (Sniffer, 2010). Generally the concentration of PM in urban areas still remains a challenge for compliance with EU standards.

1.2 Health effects and legislations for PM

The identified effects of particles upon human health include premature mortality, increased hospital admissions, allergic reactions, lung dysfunction, cardiovascular diseases and in severe cases, death (Department of Health, 2006, Health Effects Institute, 2003, Pope et al., 1995, Dockery et al., 1993). Aerosol particles have actually been identified to be efficient transport mediums for carcinogenic compounds into human lungs (Siegmann and Sattler, 1996). Health problems of greater severity are mainly observed in susceptible groups (which include children, the elderly and people with pre-existing heart and lung diseases) and are also linked with personal exposure over long periods of time to PM. Exposure to PM has been found to trigger,

or in some cases increase the severity of chronic obstructive pulmonary diseases(COPD)(MacNee and Donaldson, 2003), short term exposure to particulate matter has resulted in aggravations of respiratory symptoms and decline in lung function (Hoek et al., 1998). Fatality can also occur when pulmonary inflammations from reaction to exposure to PM lead to cardiovascular effects such as systematic inflammation resulting in an imbalance of coagulation factors leading to in the interference of the heart rhythm(Donaldson et al., 2005). Epidemiological studies have shown a link between exposure to ambient particulate matter and adverse health effects (ABBEY et al., 1999, Cifuentes et al., 2000, Dockery et al., 1993, Wichmann and Peters, 2000)however it has been unclear as to which components or size fractions of the airborne particles is largely responsible for exerting the observed toxic effect. Recently, however, a study by Atkinson et al., 2010 considering daily concentrations of particulate mass, number concentrations and particle composition against information on death and hospital admissions over a period of time using a Poisson regression time series model found that respiratory outcomes were mainly caused by non-primary $PM_{2.5}$ and other secondary pollutants, while admissions and mortality were associated with particle number concentrations. They showed that specific mixtures of particle air pollution may be relevant to specific diseases. Their study so far provides the best information available on the importance of PM mass measures to public health even though it was based upon data from a single monitoring site.

The Committee on Medical Effects of Air Pollution (COMEAP) published a report in 2010 which indicated that approximately 4 million life years would be saved or an increase in life expectancy of 20 days in people born in 2008 for each 1 $\mu g\ m^{-3}$ decrease in $PM_{2.5}$ concentration over the lifetime of the current population of England and Wales. COMEAP stated that in 2015 that 30,000 deaths in the UK could be attributable to exposure to $PM_{2.5}$ (which is about 23% of all respiratory deaths) and about 130,000 emergency admissions to hospital. which was more

than figures for passive smoking and car accidents (COMEAP,2016). The World Health Organisation rank PM as the 13th leading cause of mortality worldwide and estimate that approximately 800,000 premature deaths are contributed by PM each year (Morgan et al, 2016).

Ultrafine particles are small enough to penetrate into the lungs and potentially transfer into tissues to cause physiological damage based on their chemical complexity (Schwartz et al., 1996, Cifuentes et al., 2000). The smaller particles that penetrate deeper into the tissues, deposit in the air sacs and can cause inflammation. This can result in the change of the clotting properties of the blood leading to increase in the chances of heart attacks (Pope CA III, 2006). The schematic representation of deposition location in the human respiratory tract is shown in Figure 3.

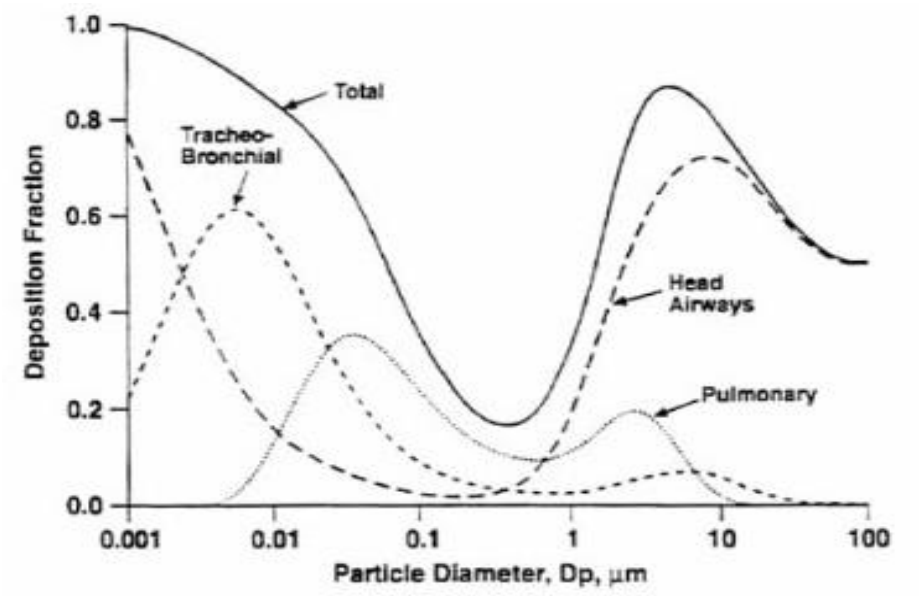


Figure 3 Particulate matter size and deposition in the human body BENSON (2012).

In 2000, researchers Jaques and Kim carried out measurement of total deposition fraction of ultrafine aerosols in a small group of young healthy adults (11 in number) and revealed that regardless of breathing patterns, the deposition fraction increased as particle size decreased (Jaques and Kim, 2000). Regardless of the study size, their findings were consistent with the

respiratory tract deposition models proposed by the International Commission on Radiological Protection (ICRP) and the U.S. National Council on Radiation Protection and Measurements (NCRP), where sub-micrometer aerosols were found to have larger deposition efficiency in the respiratory tract, especially the alveolar region (James et al., 1991, Phalen et al., 1991). The bronchial and alveolar deposition fractions was found as 0.03 and 0.16, respectively for 2 µm aerosols and this fraction increased to 0.06 and 0.28, respectively, for 0.06 µm sized aerosols. The Chemical properties of aerosols have also been found to be important characteristics as they influence the behaviour of the particles after deposition in the respiratory tract through volatility and solubility. It was found that acidic particles can be irritants when inhaled; PAHs (polycyclic aromatic hydrocarbons) are potentially carcinogenic and also any trace metals delivered to the lungs on fine particles may catalyse production of tissue-damaging oxidants (Dreher et al., 1997, Ghio et al., 1996, Hoek et al., 1998). Hoek et al., (2000) actually found association of fine particles with total mortality based on presence of sulphate, nitrate and black smoke. Svedhal et al, (2013) found that inhalation of cooking fumes slightly alters the expression of inflammatory reactions in the bronchial mucosa and its subsequent systemic inflammatory response in blood biomarkers after short-term exposure to moderate concentrations of cooking fumes. Recently Yu et al., 2015 showed that there is an incremental lifetime cancer risk resulting from exposure to the household air pollution from cooking is higher than the acceptable level of 10^{-6} making it a serious health concerns. In as much as the chemical composition is important, the inhalation of PM causes irritation to the respiratory tract when it is exposed to it.

In toxicological and controlled human exposure studies, several physical, biological and chemical characteristics of particles have been found to cause cardiopulmonary responses. Amongst the characteristics found to be contributing to toxicity in epidemiological and controlled exposure studies are metal content, presence of PAHs, other organic components,

endotoxin and both small (< 2.5 μm) and extremely small size (< 100 nm). Physical characteristics of PM are particle size, surface and number and the smaller the particle, the larger is the surface area available for interaction with the respiratory tract, and for adsorption of biologically active substances. Organic compounds are common constituents of particles, and comprise a substantial portion of ambient PM. Some of these compounds extractable from PM (especially PAHs) have been found to exert pro-inflammatory as (Li et al,2000; WHO, 2003). Some of the PAHs and their nitro-and oxy-derivatives have been shown to be mutagenic. There has also been increasing evidence that soluble metals may be an important cause of the toxicity of ambient PM. The transition metals are also important components concerning PM-induced cardiovascular effects (Clarke et al, 2000). Transition metals potentiate the inflammatory effect of ultrafine particles (Wilson et al, 2002). However, it has not been established that the small metal quantities associated with ambient PM in most environments are sufficient to cause health effects.

1.2.1 Legislation

Early epidemiological studies in USA identified PM_{10} as an important pollutant metric related health effects and it was considered so until further advancement in research identified the smaller particles within PM_{10} ($PM_{2.5}$)as being the most significant in relation to health outcomes (SNIFFER, 2010). Experts are still debating onto which fraction is primarily responsible for the health outcomes (size or a non-mass metric) (AQEG, 2012, Harrison et al., 2012).

Various governments have long since established emission standards for PM_{10} such as United States Environmental Protection Agency (USEPA), European Union (EU), UK Department for Environment Food and Rural Affairs (DEFRA); due to its negative impact on human health and poor visibility.

Initially the Air Quality Strategy for England, Scotland, Wales and Northern Ireland (AQS) set the PM_{10} annual mean of 40 μg m^{-3} and 50 μg m^{-3} for 24 hour mean: not to be exceeded more than 35 times a year. This was to be reduced to annual mean of 20 μg m^{-3} and 50 μg m^{-3} for 24 hour mean: not to be exceeded more than 7 times a year in the UK by the end of 2010 (APEG, 1999). The Air Quality Expert Group (AQEG) independently drew conclusions, in their report on UK particulate matter in 2005, that the European Union(EU) annual mean limit value set for 2005 would be met nearly everywhere, and concluded that there was likely to be substantial exceedences of 20 μg m^{-3} near to major roads in 2010(DEFRA, 2007, AQEG, 2005). The objective was then reviewed and not changed, due to the observation that PM_{10} annual and 24-hour mean values were expected to continue to exceed their specified limits well after their target achievement date at the end of 2010, and full compliance of the objectives across the whole country was not expected even after 2020(DEFRA, 2007, AQEG, 2005).

Evidence from studies about $PM_{2.5}$ and findings about the more chronic effect of long term exposure to PM, as well as the recognition of the absence of a threshold value for exposure to $PM_{2.5}$ lead to attention being given to this pollutant by various agencies, policy makers and expert groups in the UK. Defra included $PM_{2.5}$ into its Air Quality Strategy update in 2007 and the European Union introduced $PM_{2.5}$ standards in its Clean Air for Europe (CAFE) Directive in 2008 and this directive was implemented in UK legislation on 11 June 2010.

Epidemiological studies on large populations have been unable to identify a threshold concentration below which ambient PM has no effect on health. It is likely that within any large human population, there is such a wide range in susceptibility that some subjects are at risk even at the lowest end of the concentration range. Short-term epidemiological studies suggest that linear models without a threshold may be appropriate for estimating the effects of PM on the types of mortality and morbidity of main interest. This issue has been formally addressed in some studies (Daniels et al, 2000; Schwatz et al, 2000; Schwatz et al 2001). Methodological

problems such as measurement errors (Schwatz et al, 1990; Zeger et al, 2000) make it difficult to precisely pinpoint a threshold if it exists; effects on mortality and morbidity have been observed in many studies conducted at exposure levels of current interest. If there is a threshold, it is within the lower band of currently observed PM concentrations in Europe.

Exposure reduction approach was agreed to be the most effective and efficient way to maximise health benefits for particulates to ensure an overall reduction in exposure of the general population(DEFRA, 2007), which is an approach based on achieving a reduction in the overall exposure by the general population instead of concentrating at hot spots only.

The ability to achieve this exposure reduction target in order to improve human health is limited for UK regulators as there still exists a research gap between the understanding of contributions from regulated sources of $PM_{2.5}$ (such as industries and vehicles) and background concentrations. A lot of effort is on the way to provide better understanding of the various sources, pathways and health effects of $PM_{2.5}$, and the legislation that can contribute to its control(SNIFFER, 2010).

The EU Air Quality Framework Directive (2008/50/EC) and its sister directives in are currently the policy framework in use for the 12 potential air pollutants (which include NO_2, carbon monoxide and PM) which are known to have a harmful effect on human health. Table 1 lists the standards for PM in the UK.

Another aspect to look into is indoor air where sources of PM have been identified and there is inevitable long term exposure by the general population and mainly because the existing directives do not apply to indoor air quality and there is no government department responsible for the issue of indoor air quality in the UK. Globally the World health Organisation (WHO) have set a standard for $PM_{2.5}$ of 10 $\mu g/m^3$ annual mean 25 $\mu g/m^3$ 24-hour mean which is the concentrations at which increased mortality responses are expected to be observed due to air pollution from it and this standard is applied to the indoor environment (WHO, 2006). For the

mean time this is the standard used and it seems difficult for regulatory air quality legislation to be enforced for domestic households but the understanding of emissions from cooking practices will be useful to educate the general populace so as to make them of aware of the likely risks they are exposed to and enable them can make adequate choices on their cooking, ventilation and techniques to reduce those risks.

Air quality experts have pointed out that there is no recognised threshold below which negative health effects are absent as such human health can be affected by either short- and long-term exposure to $PM_{2.5}$ (SNIFFER, 2010). Information of the components of the fraction $PM_{2.5}$ is another important aspect that requires attention.

Organic compounds are a major constituent of particulate matter and thus the importance of identification and measurement of its constituents is important especially the proportion of PAHs which are ubiquitous environmental pollutants and include some of the most carcinogenic materials (IARC, 2010). They have been classed by USEPA as probable human carcinogens (B-2 pollutants) with evidence from animal studies. The limit for PAH has been set by the Occupational Safety and Health Administration (OSHA) for 0.2 mg m^{-3} (ATSDR, 1995), also the European union directive has proposed a target value of 1 ng m^{-3} B[a]P for total PM_{10} fraction content over an average calendar year (EUD, 2004) with suggestion on the assessment of the contribution of Benzo[a]pyrene (B[a]P)on ambient air.

An occupational exposure limit is an upper limit on the acceptable concentration of a hazardous substance in workplace air for a particular material or class of materials. It is typically set by competent national authorities and enforced by legislation to protect occupational safety and health. Occupational standards are standards that ensure the worker is not over exposed to harmful and toxic chemicals while at work. Outdoor standards on the other hand are standards

that provide an assessment of health effects of air pollution and thresholds for health-harmful pollution levels.

Occupational exposure limits are mainly intended to serve as guides to prevent illness or certain symptoms such as eye and nose irritation in industrial atmosphere and they use dose-response data which show the health effects of repeated exposure to one specific chemical. Outdoor air standards define levels of air quality that are judged necessary to protect public health in areas where the general public has access. The occupational standard as well as the ambient standard cannot be compared because the levels of exposure to the general public involves varying health status and age. Usually ambient air quality standards are substantially lower than occupational standards which are based on the time spent at work. Data is not available for extended low-level exposures to a combination of contaminants as is the case for indoor air quality problems.

The OSHA (Occupational Safety and Health Administration) occupational exposure standard for Particulate matter for respirable dust is 5 mg/m^3 and for total particulate matter is 10 mg/m^3 (for grain dust) and 15 mg/m^3 (nuisance dust); while the NIOSH(National Institute for Occupational Safety and Health) has a standard for total particulate matter (grain dust) of 4 mg/m^3 (Donham, 2002).

Table 1 Standard for $PM_{2.5}$ and PM_{10}(Sniffer 2010)

	Pollutant	Time Period	STANDARD	TO BE ACHEIVED AND MAINTAINED THEREAFTER
	$PM_{2.5}$	Annual mean	Objective of 25 µg/m^3	2020
UK		3 Year running	Objective of 15% reduction in	
		Annual mean	concentrations measured across	Betw een 2010 and 2020
			urban background sites	
	PM_{10}	24-hour mean	Objective of 50 µg/m^3	2005
			not to be exceeded more than 35	
			times a year	
		Annual mean	Objective of 40 µg/m^3	2005
Scottish	$PM_{2.5}$	Annual mean	Objective of 12 µg/m^3	2020
		3-year running	Objective of 15% reduction in	Betw een 2010 and 2020
		annual mean	concentrations measured across	
			urban background sites	
	PM_{10}	24-hour mean	Objective of 50 µg/m^3	2005
			not to be exceeded more than 7	
			times a year	
		Annual mean	Objective of 18 µg/m^3	2005
	$PM_{2.5}$	Annual mean	Target value of 25 µg/m^3	2010
European		Annual mean	Limit value of 25 µg/m^3	2015
		Annual mean	Stage 2 indicative limit value of 20	2020
		3 year	Exposure-reduction target relative to	2020
		Average	the AEI depending on the 2010 value	
		Exposure	of the 3 year AEI (ranging from a 0%	
		Indicator (AEI)	to a 20% reduction)	
		3 year	Exposure-concentration obligation of	2015
		Average	20 µg/m^3	
		Exposure		
		Indicator (AEI)		
	PM_{10}	24hr mean	Limit value of 50 µg/m^3	2004
			not to be exceeded more than 35	
			times a year	
		annual mean	Limit value of 40 µg/m^3	2004

The UK expert panel recommends 0.25 ng m^{-3} PAH measures as an annual average using B[a]P as an indicator of the PAH mixture. Presently this value is exceeded in areas which emit PAH such as urban areas and industrial areas with urban areas having concentrations 1-2 fold higher than in rural areas of Europe and up to 3 orders of magnitude higher than Arctic Canada(Prevedouros et al., 2004). The EU target value of 1ngm^{-3} on an annual average has also been found to be difficult to meet in the French alpine valleys (Marchand et al., 2004).

A review by Ravindra et al., in 2008 on PAHs emphasised a need to include all known probable carcinogenic PAHs in a new air quality index to ensure adequate protection of human health (Ravindra et al., 2008).

1.3 Cooking and Particulate Matter

In less economically developed countries indoor smoke exposure is a recognized major cause of ill health (Kurmi et al., 2008; Pandey et al., 1989; Smith et al., 2000; Smith, 2003). This is greatly as a result of the fact that in these developing countries the fuels for heating and cooking are primarily biomass fuels such as wood, dried cow dung and charcoal with studies showing that high indoor concentrations of PM are generated from biomass or solid fuels (Shrestha and Shrestha, 2005). Combustion of these fuel for cooking generally leads to emission of a complex mixture of particulate and gaseous species many of which are known health-damaging pollutants. Some of these pollutants contribute to high levels of commonly regulated pollutants in the ambient environment such as respirable particulate matter (PM), carbon monoxide (CO), nitrogen oxides (NOx) and sulphur oxides (SOx). Cookstoves also emit both gas and particulate phase polyaromatic hydrocarbonsand oxygenated polycyclic aromatic hydrocarbons that may mediate health impacts via the formation of proteins and DNA adducts and generation of reactive oxygen species to enhance oxidative stress (4-7). Combustion of solid fuel in inefficient cookstoves generally results in the production of these variety of health-damaging gases and particles (Smith et al. 2009), such as black carbon (BC), organic carbon (OC), methane, and carbon monoxide. As such cooking fuel will generally lead to a larger releases of pollutants making the quantity from cooking ingredients or methods very negligible as found by Chafe et al in 2014 where an energy supply–driven emissions model (GAINS; Greenhouse Gas and Air Pollution Interactions and Synergies) was used along with a source-receptor model (TM5-FASST) to estimate the proportion of ambient $PM_{2.5}$ ($APM_{2.5}$) produced by households and the proportion of household $PM_{2.5}$ emissions from cooking with solid fuels. Household cooking with solid fuels was found to account for 12% of $APM_{2.5}$ globally with observed $APM_{2.5}$ values of 37% (2.8 $\mu g/m^3$ of 6.9 $\mu g/m^3$ total) in southern sub-Saharan Africa and South Asia having the highest regional concentration of $APM_{2.5}$ from household cooking (8.6 $\mu g/m^3$)

(Chafe et al; 2014). $PM_{2.5}$ from cooking constituted more than 10% of $APM_{2.5}$ in seven regions housing 4.4 billion people. They also observed that as the countrys economic status increased there was an accompanied decrease in the contribution of household cooking to APM2.5 signifying the decrease in dependence to inefficient cooking fuels and technologies. They estimated that exposure to $APM_{2.5}$ from cooking with solid fuels caused the loss of 370,000 lives and 9.9 million disability-adjusted life years globally in 2010 on the basis of GBD 2010 (Chafe et al; 2014).

The important contribution of household fuel use (for heating and cooking) to particulate matter emissions has been established in many studies such as in China where residential coal and biomass combustion were found to be key source of fine particulate matter (≤ 2.5 μm in aerodynamic diameter; $PM_{2.5}$) which accounted for 47% (4.3 Tg of 9.3 Tg total) and 34% (4.4 Tg of 13.0 Tg total) of China's $PM_{2.5}$ emissions in 1990 and 2005 (Lei et al. 2011). In 2000, 86% of BC emissions in both India and China was attributed to residential coal and biomass use with 96% and 97% of OC emissions attributed to these fuels in India and China respectively (Ohara; 2007).

About 3 billion people (mainly poor people that live in low- and middle-income countries) still cook and heat their homes using solid fuels (i.e. wood, crop wastes, charcoal, coal and dung) in open fires and leaky stoves around the world. The indoor smoke can be 100 times higher than acceptable levels for fine particles in poorly ventilated dwellings and the exposure has been found to particularly high among women and young children, who spend the most time near the domestic fireplace (WHO,2014).

The International Agency for Research on Cancer (IARC) has concluded that emissions from household use of coal are a Group 1 carcinogen, whereas those from biomass are Group 2(a), a probable carcinogen, with more limited epidemiologic evidence further showing that fuel type contributes to the emissions generated during cooking. For lung cancer 25case studies

were investigated for household coal use, 7 of which provided cooking-specific estimates. Exposure was assessed by fuel type, and lung cancer was confirmed by pathology for most cases while for biomass and cancer 14 eligible studies of cooking and/or heating with biomass were analysed by IARC to obtain sex-specific estimates and examine exposure-response evidence and from these it was determined by fuel type and as with coal, most cases of lung cancer were confirmed pathologically (Smith et al., 2014).

A study by He et al 1991 carried out a quantitative risk assessment of indoor air pollution and found that indoor air pollution is the main risk factor in inducing lung cancer in Xuan Wei County. They found that the presence of lung cancer in females in that region was statistically significantly associated with chronic bronchitis and family history of lung cancer. The results also suggested an association of lung cancer with duration of cooking food, but not with passive smoking. Studies which are carried out in locations where solid fuels are used have been found to have elevated concentrations attributed largely from the combustion of the fuel used for cooking rather than from the cooking itself. Measured mean concentrations of 24-hr concentration of $PM_{2.5}$ in using households using solid cook fuels in India was found to range from 163 $\mu g/m^3$ in the living area to 609 $\mu g/m^3$ in the kitchen area (Balakrishnan et al., 2013). In developed countries, the use of cleaner fuel is more common as such the main contribution of exposure of emission from cooking is accounted from compounds derived from the cooking of the ingredients itself. This is discussed in further details in section 1.4 as well as shown in Table 4, Table 9, Table 5 where a list of various studies showed magnitude and concentrations contributed from cooking in countries such as USA, Taiwan, Switzerland, UK Czech Republic, showing that even high clean fuel are used, cooking does provide a change in personal dependant on the type of meal cooked or the style of cooking.

In such countries combustion of fuel such as coal, wood, peat and gas for cooking or heating, and the combustion of tobacco in the form of smoking are the main sources of PM indoors(Lai

et al., 2006; Semple et al., 2012). Indoor PAH levels usually range from 1 ng/m^3 to 50 ng/m^3 due to tobacco smoke and residential heating with wood, coal, and other materials (WHO, 1998). Environmental tobacco smoke is a major contributor to air pollution and dust, and surfaces remain contaminated long after the smoking has ceased (called third-hand smoke). Measurement of PAHs in settled household dust in 132 homes showed that total PAHs were 990 ng/g in smoking households versus 756 ng/g in nonsmoking households (Hoh et al., 2012). Cooking has actually been found to be a very significant particle generating activity indoor and this has generated lots of interest (He et al., 2004c, Hildemann et al., 1991a, Nicole., 2014; Nolte et al., 1999, Robinson et al., 2006, Rogge et al., 1997, Schauer et al., 1999b). Numerous studies that have been carried out around the world have related adverse health effects to practices of cooking and also from the products of incomplete combustion. There are very few meals that can be eaten without cooking such as salads and sushi. Foods like meats, grains and vegetables cannot be eaten raw and have been identified to be prepared using varying culinary techniques based on regions of the world it is being prepared. Differences exist spanning from the spices used, to differences in fuel type, to cooking duration among other things.

Over the years networks of restaurants and cooking kiosks have sprung up all over cities to cater for the population of busy people who need to eat during their scheduled days. As a result of this emissions from these cooking establishments tend to emit exhausts from the processes onto streets and surrounding areas. A clear understanding of what exactly is released into the air is important and essential for understanding if there might be any negative health effects as a result of these emissions as this an unregulated air pollution source.

In cases of people who mainly eat freshly prepared homemade food, these meals are mainly prepared indoors by themselves or other family members, as such, finding out how much these cooking practices also affects indoor air cannot be overemphasized.

Household air pollution exposure was linked to 3.5 million deaths with an additional 0.5 million deaths from ambient/outdoor air pollution resulting from solid fuel use, in the Global Burden of Disease/Comparative Risk Assessment Project (GBD 2010) report published in 2012(Lim et al., 2012). Estimates from the WHO for 2012 (WHO 2014), relying on an updated methodology, estimated that the global mortality burden of household air pollution at 4.3 million people annually. Worldwide more than a million people die from chronic obstructive pulmonary disease (COPD) annually due to indoor exposure to smoke which generally contains a range of health-damaging pollutants, such as fine particles and carbon monoxide (Hetland et al., 2000). The use of solid fuels (biomass and coal) for cooking and heating homes is practised by around 3 billion people in open fires and leaky stoves, especially by people with low and medium resources in developing countries. As such, poorly ventilated homes can have indoor smoke concentrations of respirable particles of more than 100 times the acceptable levels (Hetland et al., 2000; Kurmi et al, 2008) with mostly women and young children being exposed to these extremely high levels. Extensive literature on the harmful effects of cooking with solid fuels have been made over time. The GBD and WHO estimate draws on epidemiological literature that links indoor fine particulate matter (PM) emissions from solid fuel combustion with acute lower respiratory infections, chronic obstructive pulmonary disease (COPD), lung cancer, cataracts and low birth weights (Dherani et al., 2008; Ezzati and Kammen, 2002; Kurmi et al., 2010.; Pokhrel et al., 2005; Smith et al., 2004). The estimates are believed to be underestimated as the values exclude the full health impacts of households cooking with unprocessed solid fuels on asthma; tuberculosis; childhood nutritional deficiencies, including anemia and stunted growth; blindness; maternal depression; cognitive impairment in the young and old; upper respiratory, digestive, and cervical cancers.

A study of domestic workers in rural Nepal found that they were exposed to average respirable dust concentrations of 1400 µg /m^3 which is more than the current UK limit for respirable dust

(4000 µg/m^3). High respirable dust concentrations and exposures are thus likely to produce respiratory illness to the Homemakers who spend a large proportion of their lives indoors in these. Exposure can be controlled by the use of different fuel types and/or the use of flued stoves.

Indoor levels of particles in developed countries are much lower than in developing countries and this is generally attributable to the advancement in technology for general household activities and also the use of cleaner fuels (such as liquefied petroleum gas, electricity and natural gas) for cooking and heating. However, there are still observed risks to health in people exposed to indoor air in these locations. Legislation relating to air pollutant exposure in developed countries is normally based upon ambient outdoor concentrations, potentially leading to inadequate protection of the general public who spend the majority of their time at home, offices or other enclosed locations where the concentrations of some pollutants are often much higher than ambient levels (Marcazzan et al., 2001). Knowledge of the indoor environment is limited and is of great importance as the majority of people have been found to spend about 80-90% of their time indoors in many countries (Scapellato et al., 2009, Koistinen K.J., 2001, Delgado-Saborit et al., 2011). Also the indoor environments have been found to be affected by factors such as the design of the buildings, insulation and ventilation in order to ensure an adequately controlled environment for thermal comfort, which can also affect level of individual exposure (Tan et al., 2012). The level of exposure from cooking emission are generally affected by certain factors which can include things like the fuel type used, cooking techniques, home ventilation, ingredients used during food preparation and kitchen location. The general population is exposed to cooking-related risk regardless of race, age, wealth and cultural food preferences as cooking is an important aspect of human culture (Kim et al., 2011). There is generally a big gender difference for exposure to cooking in many countries as women tend to do most of the cooking and by this children are similarly exposed as a good number of

them are with their mothers during cooking and these group of people tend to spend longer periods of time at home (Balakrishnan et al., 2015). As such women have been found to experience higher personal exposure levels than men and therefore higher relative risk to develop adverse health outcomes due to their greater involvement in daily cooking activities (Smith et al 2014). Evidence from several countries shows that female cooks are exposed to significantly higher particulate matter emissions than men, up to four times men's levels in Kenya and up to double the level of men in South Asia studies (Dasgupta et al., 2006; Balakrishnan et al., 2004; Ezzati & Kammen., 2002; Smith et al., 2007). With some studies from Senegal, Ghana, and Peru, demonstrating evidence of greater incidence of respiratory illness and eye disease in women in solid fuel-using households based on self-reported household data (in Peru respiratory illness symptoms were30% in women and 22% in men; in Ghana 74% in women and 13% in men) (Adrianzen, 2011; Bensch & Peters, 2012; Odoi, 2010); The processes used in cooking such as frying, roasting, grilling, boiling and broiling contribute to pollutant emissions and are affected by ingredients, recipes and procedures, fuel types, temperature and extraction/ventilation equipment (Zhang et al., 2010). Table 2 summarises the cooking styles, ingredients and oils used for some common cultural culinary techniques.

Cooking contributes particles to outdoor as well as indoor air. Commercial cooking emissions may have contributed to the exceedance of the Federal $PM_{2.5}$ air quality standards in certain regions such as Pittsburgh, Pennsylvania, where meat charbroiling was shown to contribute to carbonaceous PM by Cabada et al. (2002). Commercial cooking has been identified to be an important contributor to secondary organic aerosols (SOA) condensation of gaseous organic emissions following photochemical processes), organic carbon(OC) and elemental carbon(EC) in the urban environment (Roe et al., 2005). Rogge et al. (1991) reported that 21% of the primary fine organic aerosols in the Los Angeles area in the 1980s were generated by charbroiling and meat cooking activities which was in agreement with previous studies in the

area (Hildemann et al., 1991b). A similar study in 1997 Denver Colorado, the Northern Front Range Air Quality Study (NFRAQS), found that meat cooking contributed about 15% of $pm_{2.5}$ organic aerosol concentrations (Watson et al., 1998).

Recently a study in New York City (NYC) using a High-Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS) identified that cooking and traffic were two distinct and mass-equivalent Primary Organic Aerosol sources, contributing 30% of the total Organic Aerosol (OA) mass collectively during the period (Sun et al., 2011). The average mass concentration of Cooking OA was 1.02 $\mu g\ m^{-3}$ which was higher than the mass concentration of Hydrocarbon like OA (0.91 $\mu g\ m^{-3}$), which was surprising as the sampling site was actually close to two major highways (<1 mile), giving a clear indication that cooking activities were an important source of primary particles in NYC.

A prior air quality campaign at Beijing in 2008 found that 24.4% of total organic mass was similarly attributed to cooking related organic aerosols, with a similar use of a HR-ToF-AMS (Huang et al., 2010) .

Measurement of particle number and size distribution of particles generated during cooking has been carried out in various studies to provide a better understanding of characteristics of particles generated during cooking (Abt et al., 2000; Buonanno et al., 2009;Dennekamp et al., 2001; See and Balasubramanaian., 2006a; Wallace et al., 2004; He et al., 2004a).

Several studies have shown evidence of adverse effects on human health from cooking emissions (Ko et al., 2000, Yu et al., 2006). An association was found between lung cancer and oil fumes from Chinese cooking in non smoking Taiwanese women in China by Ko et al., (2000). Lung cancer risk was found to be increased with the number of meals per day to about threefold for women who cooked these meals each day. They ascribed this finding mainly to the frying of ingredients in oil in Chinese cooking, which produces plenty oil fumes to which the cooks were exposed. Higher risk of occurrence was observed in women who wait until the

oil reached a high temperature before cooking the foods and those that do not make use of a fume extractor.

Table 2 General cultural styles of cooking and common ingredients, oils and spices used during cooking.

COOKING STYLE	METHOD	INGREDIENTS	OIL	SPICES
Chinese	Stir fry, simmer, steam roast stew	MAIN- Meat type- Pork, sea food, poultry, beef, Vegetable-cabbage, carrots, cucumber, broccoli OTHERS- Eggs, ginger, hot pepper, scallion, garlic, rice, flour, peanuts, fruits	Soy beans Peanut oil Canola oil	essence of chicken, salt, peanut oil, light soy source, sugar
Western	Grill, broil, roast, deep fry, stew,	MAIN -Meat type-beef, chicken Vegetables-carrots, broccoli, OTHERS - milk, flour	Corn oil, vegetable oil olive oil,	Salt, black pepper, garlic, basil, parsley
Fast Food	Deep fry, stew	MAIN - Meat type- beef, chicken, Potatoes	Vegetable, butter, corn oil	Salt
African	Deep fry, boiling, stew	MAIN- Meat- beef, chicken, fish Vegetables-spinach OTHERS- yam, rice, plantain, banana.	Ground nut oil, palm oil, vegetable oil	Thyme, curry
Indian	Deep fry, boiling, stew	MAIN- Meat- fish and chicken OTHERS- rice, flour, beans, lentils, pearl millet, wheat flour, milk, yoghurt, plantain. .	Vegetable oil, peanut oil, mustard oil, coconut oil, sesame oil,	Chilli pepper, black pepper, mustard seed, cumin, turmeric, ginger, cardamom, cinnamon, clove, garam masala, coriander, garlic, mustard seeds, nutmeg, mint
Malay	Deep fry, boiling, stew	MAIN- Meat-Fish, squids, prawns, crabs , chicken, beef and mutton. OTHERS-rice, noodles, yoghurt, coconut milk.	Vegetable oil, coconut oil, sesame oil,	Lemongrass, shallots, ginger, chillies, garlic, turmeric, lime leaves, laksa leaves, wild ginger flower buds or torch ginger and screwpine leaves, fennel, cumin, coriander, cardamom, cloves, star anise, mustard seeds, and nutmeg

Metayer (2002) also observed an elevated incidence of cancer among non-smoking women who had long term exposure to cooking fumes (Metayer et al., 2002). A territorial-wide survey

in Hong Kong by To et al., (2007), found that the oil fumes collected from exhausts of 15 restaurant kitchens sampled, contained carcinogenic compounds such as PAHs and aromatic amines and some gas-phase aliphatic hydrocarbons(To et al., 2007). These compounds generated from the fumes emitted from the cooking oil combustion during the process of stirring or deep-frying can condense on the surfaces of particles. The exposure to carcinogenic compounds released from the cooking were likely causes of the high incidence of cancers in cooks as identified.

Another analysis of respiratory illnesses in preschool children in Hong Kong revealed that household gas cooking was associated with respiratory illnesses and a dose-response relationship was observed between the frequency of gas cooking and occurrence of respiratory illnesses in area with relatively low outdoor air pollution. The health impact of gas cooking on the respiratory health of the subjects was assumed to also have resulted from the exposure to cooking fumes and also nitrogen dioxide (Wong et al., 2004).

Generally the risk associated with cooking is still poorly understood as such awareness is necessary to ensure adequate protection of health for the general public.

1.4 Emissions from cooking

Studies of cooking emissions have been carried out in both real life kitchens and in controlled environments. It is assumed that in controlled experimental setups, the measurements are influenced mainly by the fuel used and the food being cooked while in actual real life kitchens measurement of emissions are influenced by many factors such as room arrangement, building materials, outdoor infiltration, other combustion devices, ventilation, and cooking methods (Huboyo et al., 2011).

Visible fumes are generated during the cooking process, which are usually due to submicrometer sized particles, which consist of oil droplets, combustion products, steam from water in the food being cooked and condensed organic pollutants. The particulate matter (PM)

generated is generally within the ultrafine particle (UFP) - which represents particles of diameter less than100 nm - and fine PM ($PM_{2.5}$) size ranges. The physical stirring of food has been found to lead to the generation of large aerosols due to the process of splashing of the ingredients (Long et al., 2000). The combustion process associated with cooking can lead to the formation and direct emission of ultrafine particles (UFP) to the atmosphere, and hot vapours in the cooking fumes may also cool and nucleate to form more UFP (Sioutas et al., 2005, Lai and Ho, 2008). These particles may contain organic substances, such as polycyclic aromatic hydrocarbons (PAH) and heterocyclic amines, adsorbed on their surfaces (Ho et al., 2002).

There is a scarcity of national inventories of cooking activities, but an attempt was made by Roe et al. (2005) to compile a national emission inventory for commercial cooking in the United States as listed in Table 3. For comparison, data for highway vehicles extracted from the National Emissions Inventory (NEI) Air Pollutant Emissions Trends Data for the same year (Chappell et al., 2003), show that although traffic emits orders of magnitude more CO and VOC than cooking, particulate matter emissions from cooking are comparable with those emitted from highway vehicles. This is consistent with a study of Li et al. (2003), who found that the emission rates of total PAH from cooking sources in the study city (i.e. emissions from both restaurants and home kitchens), were slightly lower than those for traffic sources in a representative city of Taiwan (8,973 kg/year for cooking against 13,500 kg/year for traffic). Nonetheless, they observed that the emission rate for $B[a]P_{eq}$ toxic equivalent for cooking sources was much higher than that from traffic sources (675 kg/year from cooking and 61.4 kg/year emitted from traffic sources). This indicated that cooking PAH may cause much more serious problems than traffic sources in terms of carcinogenic potency (Li et al., 2003).

Table 3 National emissions rate (tonnes/year) of criteria pollutants from commercial cooking in the USA (Roe et al., 2005) and for highway vehicles (Chappell et al., 2003)

Pollutant	Total charbroiling	Deep frying	Flat griddle frying	Clamshell griddle frying	Under-fired charbroiling	Conveyorized charbroiling	Highway vehicles
VOC	115	1,170	39	940	7,200	2,100	4,400,000
CO	33,000		1,900		23,700	7,400	48,400,000
$PM_{2.5}$	79,300		11,900	910	58,300	8,200	135,000
PM_{10}	85,500		15,700	1,100	60,300	8,500	192,000
PAH total	206		41		122	43	

1.5 Particle mass concentration

The PTEAM Study (Particle Total Exposure Assessment Methodology) performed in the US, reported around 20 $\mu g/m^3$ higher particle concentrations in houses where cooking took place during their monitoring than those house where no cooking occurred (Wallace, 1989). They reported that the proportion of $PM_{2.5}$ and PM_{10} due to cooking represented 25% for both particle sizes. This proportion increased to 65% and 55%, respectively, when considering indoor sources alone (Ozkaynak et al., 1996). The subjects selected in the study were found to have high PM concentration which was finally attributed to personal cloud made up of PM matter from cooking, cleaning or living with a person that smokes. Source apportionment of PTEAM ambient and personal exposure samples using a combined receptor model found that cooking was the largest contributory source of PM indoors, responsible for about 52.5% of the personal exposure samples and 43.2% of residential indoor concentrations (Zhao et al., 2006). After 1,000 hours of cooking, they also found that the mean $PM_{2.5}$ personal exposure increased an average of 56 $\mu g/m^3$ while cooking activities took place, and that cooking increased the overall 24-hours personal exposure about 2.5 $\mu g/m^3$ in those persons that had cooked during the sampling day (Wallace et al., 2006).

A study to characterize indoor sources of particles conducted in Boston, USA, made measurements of particle size and volume concentration over 6 days in four non-smoking households equipped with gas and electric stoves (Abt et al., 2000). The monitoring equipment was placed in a single indoor location adjacent to the kitchen and living room and from the data obtained, it was found that the highest mean peak mass concentrations were for barbequing and sautéing for the $PM_{0.02-0.5}$ and $PM_{0.7-10}$ respectively, whilst the lower mean peak concentrations were found for frying and oven cooking or toasting for the same size ranges respectively (Abt et al., 2000) as shown in Table 4 .

Another US study found that the average $PM_{2.5}$ concentration due to cooking over 195 cooking events was about 5.5 µg/m^3 with a standard error of 2.3 µg/m^3 (Allen et al., 2004). In Europe, a study made a comparison of elderly residents in Amsterdam (47) and Helsinki (37), and found that the estimated contribution from cooking ranged from 1.9 µg/m^3 for indoor $PM_{2.5}$ in Helsinki to 3.4 µg/m^3 for $PM_{2.5}$ personal exposure concentrations (Brunekreef et al., 2005).

Rates of emission of aerosol have been reported to vary based on type of appliance used, the cooking conditions used and fat content of meat (McDonald et al., 2003). In an experiment where hamburger, steak and chicken were grilled and charbroiled, McDonald et al. (2003) found that the $PM_{2.5}$ emission rate for charbroiling meats ranged between 4.4 to 15 g/kg. The largest quantity of $PM_{2.5}$ was emitted by hamburger (15 g/kg) which had higher fat content (30%) and were cooked on a char broiler. These results are consistent with data reported by Hildemann et al. (1991a). McDonald et al. (2003) reported that charbroiling produced higher concentrations than frying, 12-46 g/kg meat when charbroiling vs. 0.57 g/kg meat when frying. They also reported that charbroiling lean meat produced less concentrations of particles in the smaller size range (<20 nm) and in the larger size range (>100 nm) than regular meat.

Similarly, Buonanno et al. (2009) found that gas stoves emitted more particles than an electric stove when frying resulting in higher indoor concentrations when gas stoves were used (60-

118 μg/m^3) than when electric stoves were employed (12-27 μg/m^3); and that emission rates were considerably affected by the type of food used such as listed in Table 2. Increased emissions measured at the source were reported to be a function of increased cooking temperature. Foods containing a higher percentage of fat generated higher emission rates than those with less fat percentage. They reported higher aerosol mass emission when cooking fatty foods resulting in higher indoor concentrations (280-389 μg/m^3) than when cooking vegetables (78 μg/m^3). Particle emission factor varied significantly also with type of oil used. Sunflower oil generated the lowest mass emission factors, whilst the highest emissions were from olive oil (Buonanno et al., 2009). Glytsos (2010) reported that frying of onions in olive oil in a controlled room emitted $PM_{2.5}$ increasing the indoor concentration in the range of 70 to 600 μg/m^3 (Glytsos et al., 2010).

Several studies have found that Asian style cooking emits more particulate matter than Western cooking with concentrations of $PM_{2.5}$ ranging 30 to 1,400 and 20 to 535 μg/m^3 as reported by various groups (Lee et al., 2001a, Levy et al., 2002, He et al., 2004c)

A summary of the main studies reporting aerosol concentration emitted from cooking and the reported concentrations can be found in Table 4.

1.6 Particle size distribution

The size distribution of aerosols emitted from cooking activities has been reported in several studies whose methodology and study description is summarised in Table 5 and results are compiled in Table 6. Generally some of these studies have shown that indoor particle concentrations are substantially affected by cooking activities, cleaning and the movement of people (Abt et al., 2000, Diapouli et al., 2011, He et al., 2004a). The largest percentage of the measured particles are ultrafine particles (UFP), with modes in the number distribution reported generally in the range of 20 to 100 nm as shown in Table 6.

He et al. (2004a) studied 15 homes in Australia while cooking was carried out under good and poor ventilation for 48 hours. They found that some indoor activities led to an increase in indoor particle number concentration of about 1.5-27 times concentrations in comparison with the particle number concentration when no indoor source was in operation. They also found an emission rate ranging 0.2-4x10^{12} particles/min and peak submicron number concentrations for cooking of 16,000 and 180,000 part/cm^3 (He et al., 2004a).

An investigation of the size distribution of particles emitted from cooking was carried out using a scanning mobility particle sizer (SMPS) in a domestic kitchen using five different cooking methods, such as steaming, boiling, stir-frying, pan-frying, and deep-frying. Deep-frying was found to have the highest particle number concentration, whilst steaming produced the lowest particle number concentration. Their observations found that cooking activities using oil produce higher concentrations than those using water (See and Balasubramanian, 2006a). They reported a 24-fold increase in particle concentration observed between deep frying and background concentrations (6.0×10^5 cm^{-3} compared to background concentrations which were 2.5×10^4 cm^{-3}) (See and Balasubramanian, 2006a). In another study, they characterised Chinese cooking emissions, and found that the average number concentration increased by factor a of 85 during the cooking periods (7.7x10^5 part/cm^3 compared to 9.1x10^3 part/cm^3 during non-cooking hours (See and Balasubramanian, 2006b).

Yeung and To (2008) examined aerosols generated by commercial food preparation and found a lognormal size distribution. Increased cooking temperature resulted in an increased modal diameter of aerosols. Higher cooking temperature also increased the normalized number concentration sub-micrometer aerosols (between 0.1 and 1.0 μm) (Yeung and To, 2008).

Siegmann and Sattler (1996) found that diameter and number concentrations of oil droplets increased with an increase in temperature. They analysed aerosols from different hot vegetable oils and obtained a size distribution with a mean droplet size range of 30 nm at 223ºC to 100

nm at 256ºC. Particle number concentration increased from 2.25 x 10^5 part/cm^3 to 4.5 x 10^5 part/cm^3 in the same range of temperatures (Siegmann and Sattler, 1996).

Dennekamp et al. (2001) studied the generation of ultrafine particles and nitrogen oxides using different cooking procedures comparing gas and electric stoves in a laboratory. They found higher concentrations of particles in the size range of 15-40nm (and also oxides of nitrogen) when cooking on gas (Dennekamp et al., 2001). The smaller particles generated were found to grow in size with time during the experiment. The high concentrations of pollutants observed were attributed to the absence of ventilation in their laboratory kitchen.

Frying of onions in olive oil in a controlled room to characterise contributors of particle concentrations in indoor environments produced high particle concentrations, ranging between 9 - 15 × 10^4 particles cm^{-3} (Glytsos et al., 2010). High emission of nanoparticles were reported during frying (1.15 x 10^5 part/cm^3, mainly 20 nm). However, sometime after the frying stopped (i.e. 45 min later), the number concentration decreased down to 4 x 10^5 part/cm^3 and particles become larger leading to a bimodal size distribution indicating a strong coagulation effect (Glytsos et al., 2010), which is consistent with previous studies (Sjaastad et al., 2008, Dennekamp et al., 2001).

A study in an apartment in Taiwan found a range of mode diameters of particles concentrations between 30-50nm for domestic cooking processes of scrambling eggs, frying chicken, and cooking soup with higher mode diameter for frying chicken (Li et al., 1993). Similarly, in an 18 month campaign in a four bedroom house consisting of three levels located near Washington DC, USA; particles generated from cooking were found to be mainly in the ultrafine range (about 90% of total particles), with frying being found to generate more particles than any other cooking method (Wallace et al., 2004), consistent with recent studies (Buonanno et al., 2011, Huboyo et al., 2011, Hussein et al., 2006).

Buonanno et al. (2009) sought to evaluate the influence of temperature, oil, food and stove type on particle number, surface area and mass emission factors consequence of cooking with different methods such as grilling and frying. They used a Scanning Mobility Particle Sizer (SMPS) and Aerodynamic Particle Sizer (APS). They found that frying food with oil using an electrical frying pan produced emission factors well below those observed for frying using a gas stove. The particle emission factor was also dependent upon the temperature of the stove, with values 9 and 4 times higher at the maximum stove power for gas and electric stoves respectively (Buonanno et al., 2009), consistent with previous studies of Siegmann and Sattler (1996), Dennekamp et al. (2001), Yeung and To (2008) and To and Yeung (2011). In another study by them, they reported high particle indoor concentrations (3×10^4–6×10^5 particles cm^{-3}) in 14 pizzerias and PM_1 concentrations of about 10–327 $\mu g\, m^{-3}$ during normal ventilation conditions (Buonanno et al., 2010). However most of the particles generated in this study are believed to be from the wood burning used to fire the oven, and highlights the high particle concentrations that can build up in such microenvironments. In another study, Buonanno and colleagues found that frying the same type of food consistently emitted more particles than grilling, with a factor of 1.4-1.5 (Buonanno et al., 2011).

1.7 Organic compounds emitted during cooking

Cooking involves a wide range of chemical reactions. For instance, many sugars (e.g. disaccharides or oligo-saccarides) or carbohydrates undergo hydrolysis when heated with water. The hydrolysis reaction breaks down the complex sugar into single ring sugars. If sugars are heated further, degradation reactions will occur and the sugar rings will open up to form new molecules such as acids and aldehydes. If the temperature is increased sufficiently, the degradation products may recombine to form chain-like molecules (Barham, 1950). In meat cooking, fat which occurs as triglyceride (i.e. fatty acids esterified to a glycerol backbone) in uncooked meat is hydrolysed or thermally oxidized and produces free glycerol, free fatty acids

and mono and diglycerides as shown in Figure 4 (Nolte et al., 1999). The chemical reactions that occur between proteins and carbohydrates or sugars during cooking are known as the Maillard reactions. These involve initial degradation to amino acids and smaller sugars. The acids and aldehydes produced after the opening of the sugar rings react with the amino acids to produce a wide range of chemicals (e.g. furanones) (Barham, 1950).

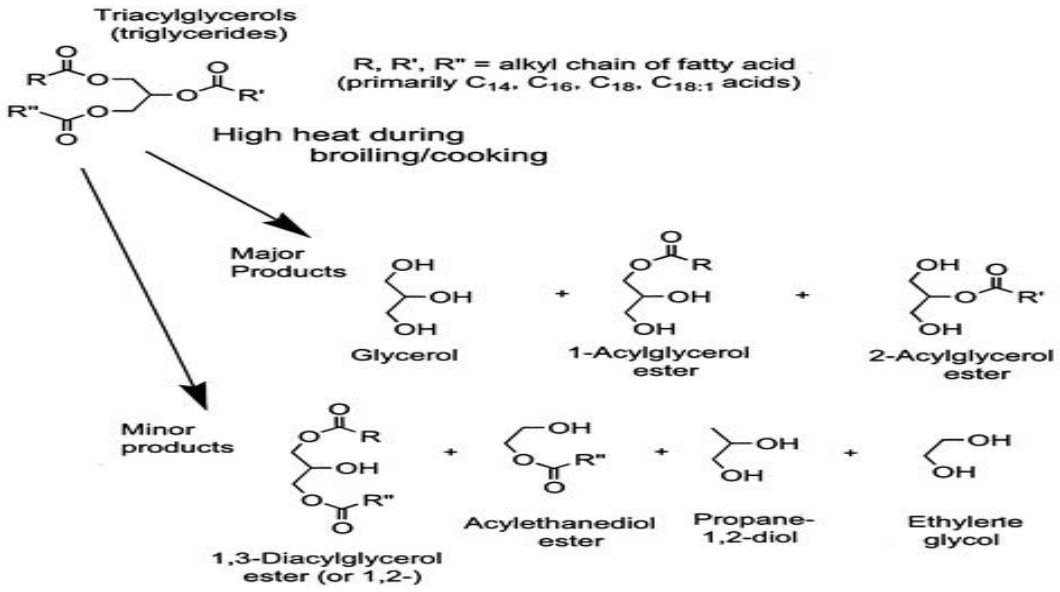


Figure 4 Break down products of triglycerides (Nolte et al., 1999)

The chemical properties of the aerosols generated during cooking can be measured to further provide useful information on the aerosol composition. In most of the studies aimed at performing chemical speciation of the cooking aerosol, samples are collected on filters for gravimetric determination and to allow subsequent chemical analysis. In some cases, denuders are used to collect the vapour phase of semi-volatile components for further analysis. Off-line chemical characterisation studies often employ sampling methods which have the potential to cause positive artefacts associated with the reaction of trace gases with particles on the filter or the filter itself. Negative artefacts may also arise from evaporative loss of semi-volatile

components. Strict sampling procedures and guidelines should keep these artefacts to a minimum (Harrison and Yin, 2005).

Table 7 gives details of key studies that have sampled and subsequently analysed the chemical composition of aerosols from cooking. Most of the off-line chemical characterisation has been done using a GC-MS analytical stage for organic speciation of the cooking emissions, with many compounds of interest requiring derivatisation. A summary of specific groups of compounds emitted from cooking identified and characterised by these studies appear in Table 8.

Table 4 Particle mass and number concentration measured in indoor environments close to cooking activities

Reference	Location	Comment	Concentration (µg/m^3)	Particle Number concentration (part/cm^3)
Li et al., 1993	Taiwan	Chicken		1.2-2.6 x10^5
Siegmann and Sattler, 1996	Switzerland	Rapeseed Oil		2.5-4.5 x10^5
Abt et al. 2000	US	Frying - $PM_{0.02-0.5}$	29	
		Frying – $PM_{0.7-10}$	19	
		Barbequing - $PM_{0.02-0.5}$	57	
		Barbequing - $PM_{0.7-10}$	12	
		Oven cooking - $PM_{0.02-0.5}$	50	
		Oven cooking – $PM_{0.7-10}$	8	
		Sauteing - $PM_{0.02-0.5}$	42	
		Sauteing – $PM_{0.7-10}$	294	
		Toasting - $PM_{0.02-0.5}$	45	
		Toasting – $PM_{0.7-10}$	8	
Dennekamp et al., 2001	UK	Frying vegetables (500 g) – gas stove		1.4 x10^5
		Frying bacon (4 racers) – gas stove		5.9 x10^5
		Frying vegetables (500 g) – electric stove		0.11 x10^5
		Frying bacon (4 racers) – electric stove		1.6 x10^5
		Bake cake – gas oven		0.9 x10^5
		Bake cake – electric oven		0.3 x10^5
		Roast meat and potatoes – gas oven		1.2 x10^5
		Roast meat and potatoes – electric oven		0.2 x10^5
		Toast –gas grill		1.4 x10^5
		Toast – electric grill		1.4 x10^5
Lee et al. 2001	China	$PM_{2.5}$ Chinese hot pot restaurant	81	
		$PM_{2.5}$ Chinese dim sum restaurant	28.7	
	Hong Kong	$PM_{2.5}$ Western Canteen	21.9	
Levy et al. 2002	USA	$PM_{2.5}$ food court	200	1.4 x 10^5
Wallace et al., 2004	USA	Cooking dinner		1.3 x10^4
		Cooking breakfast		5.7 x10^3

Table 4 Cont. Particle mass and number concentration measured in indoor environments close to cooking activities

Reference	**Location**	**Comment**	**Concentration (µg/m^3)**	**Particle Number concentration (part/cm^3)**
He et al., 2004a	Australia	$PM_{2.5}$ (48h) cooking	37	1.27×10^5
		$PM_{2.5}$ (48h) cooking pizza	735	1.37×10^5
		$PM_{2.5}$ (48h) frying	745	1.54×10^5
		$PM_{2.5}$ (48h) grilling	718	1.61×10^5
		$PM_{2.5}$ (48h) kettle	13	1.56×10^4
		$PM_{2.5}$ (48h) microwave	16	1.63×10^4
		$PM_{2.5}$ (48h) oven	24	6.15×10^4
		$PM_{2.5}$ (48h) stove	57	1.79×10^5
		$PM_{2.5}$ (48h) toasting	35	1.14×10^5
		$PM_{2.5}$ residential kitchen	535.4	2.86×10^4
He et al., 2004c	China	$PM_{2.5}$ Hunan restaurant	1406	
	China	$PM_{2.5}$ Cantonese restaurant	672	
See and Balasubramanian, 2006a, See and Balasubramanian, 2008	Singapore	$PM_{2.5}$ Steaming	66 ± 7.6	5.4×10^4
		$PM_{2.5}$ Boiling	81 ± 9.3	6.9×10^4
		$PM_{2.5}$ Stir-Frying	120 ± 13	9.3×10^4
		$PM_{2.5}$ Pan-Frying	130 ± 15	11×10^4
		$PM_{2.5}$ Deep-Frying	190 ± 20	59×10^4
See and Balasubramanian, 2006b	Singapore	Stir-fry in a wok typical Chinese food commercial food stall $PM_{2.5}$	286	7.7×10^5
See et al., 2006	Singapore	$PM_{2.5}$ Chinese stall	202 ± 141	
		$PM_{2.5}$ Malay stall	245 ± 77	
		$PM_{2.5}$ Indian stall	187 ± 44	
		$PM_{2.5}$ Background	29 ± 8	
Hussein et al., 2006	Czech Republic	Cooking in a stove, frying, oven		$0.6\text{-}1.8 \times 10^5$
Sjaastad et al., 2008	Norway	Frying Beefsteak		$1.2 \times 10^{3\,(a)}$
Yeung and To, 2008	Hong Kong	Frying vermicelli with beef		89×10^5
		Pan-frying steaks		8.5×10^5
		Pan-frying chicken fillets		8.5×10^5
		Pan –frying pork chops		8.8×10^5
		Hot oil test		6.4×10^5

Table 4 Cont. Particle mass and number concentration measured in indoor environments close to cooking activities

Reference	**Location**	**Comment**	**Concentration (µg/m^3)**	**Particle Number concentration (part/cm^3)**
Buonanno et al., 2009	Italy	Grilling in a gas stove at maximum power Cheese	283	1.1 x 10^5
		Wurstel sausage	352	1.3 x 10^5
		Bacon	389	1.0 x 10^5
		Eggplant	78	1.2 x 10^5
		Frying 50 g of chips in a gas stove at maximum power with Olive oil	118	1.2 x 10^5
		Peanut Oil	68	1.2 x 10^5
		Sunflower Oil	60	1.1 x 10^5
		Frying 50 g of chips using an electrical pan with sunflower oil	12	1.4 x 10^4
		olive Oil	27	2.6 x 10^4
		peanut Oil	13	1.5 x 10^4
Buonanno et al., 2010	Italy	PM_1 range	10-327	1.1-9.8 x 10^5
		$PM_{2.5}$	12-368	
		PM_{10}	15-482	
Buonanno et al., 2011	Italy	Grilling 100 g cheese		1.8 x10^5
		Frying 100 g cheese		2.8 x10^5
		Grilling 100 g bacon		2.0 x10^5
		Frying 100 g bacon		2.8 x10^5
		Grilling 100 g pork meat		1.6 x10^5
		Frying 100 g pork meat		2.3 x10^5
		Grilling 100 g eggplant		1.6 x10^5
		Frying 100 g eggplant		2.3 x10^5
		Grilling 100 g chips		1.5 x10^5
		Frying 100 g chips		2.3 x10^5
		Grilling 100 g onion		1.6 x10^5
		Frying 100 g onion		2.4 x10^5
Glytsos et al. 2010	Czech Republic	Frying a slice of onion with olive oil – electric griddle		1.2 x10^5

Table 4 Cont. Particle mass and number concentration measured in indoor environments close to cooking activities

Reference	Location	Comment	Concentration (µg/m^3)	Particle Number concentration (part/cm^3)
Huboyo et al., 2011	Japan	Tofu boiling	22.8 (1.21-294)	6.8 x10$^{2\,(a)}$
		Tofu frying	41.2 (1.76-707)	3.0 x10$^{2\,(a)}$
		Chicken boiling	30.8 (5.36-1,082)	2.5 x10$^{2\,(a)}$
		Chicken frying	101.6 (1.67-1,366)	1.1 x10$^{2\,(a)}$
To and Yeung, 2011	Hong Kong	Frying vermicelli with beef – gas cooking (Domestic kitchen) – PM_{10}	1,330	
		Frying vermicelli with beef – electric cooking (Domestic kitchen) – PM_{10}	1,030	
		Pan Frying of meat – gas cooking (Domestic kitchen) – PM_{10}	1,020	
		Pan Frying of meat – electric cooking (Domestic kitchen) – PM_{10}	520	
		Deep frying of chicken wings – gas cooking (Domestic kitchen) – PM_{10}	890	
		Deep frying of chicken wings – electric cooking (Domestic kitchen) – PM_{10}	680	
		Deep frying of tofu – gas cooking (Commercial kitchen) – PM_{10}	4,720	
		Deep frying of tofu – electric cooking (Commercial kitchen) – PM_{10}	3,980	
		Griddle frying of meat – gas cooking (Commercial kitchen) – PM_{10}	2,260	
		Griddle frying of meat – electric cooking (Commercial kitchen) – PM_{10}	2,600	

(a) Particles with diameter 0.3 µm <Dp<0.5 µm

Table 5 Size distribution studies for cooking aerosols

Study and Country	Location and duration	Sampling method (a)	Food	Environmental condition
Hildemann et al., 1991a USA	Commercial scale kitchen Sampling port located above the cooking surface, below the extractor fan.	Electrical Aerosol Analyser TSI 3030	Meat cooking during frying and charbroiling extra-lean and regular hamburger meat	Mechanical ventilation
Li et al., 1993 Taiwan	Domestic kitchen with a gas stove Sampling ports 3m away from the gas stove	DMA TSI 3932; CPC TSI 3022	scrambling eggs, frying chicken, and cooking soup	Windows and doors were closed during measurements
Siegmann and Sattler, 1996 Switzerland	Laboratory kitchen Hot oil at 223, 236 and 256^{o}C.	SMPS	Rapeseed oil	Closed window
Abt et al., 2000a, b USA	Domestic kitchen with gas and electric stoves. Samples collected over 6-day periods Equipment located in an indoor location adjacent to the kitchen.	SMPS TSI 3934; Electrostatic classifier TSI 3071A; CPC TSI 3022a; APS TSI 3310A	Frying, sautéing, barbequing, oven cooking and toasting	Open doors
Dennekamp et al., 2001 UK	Laboratory kitchen with gas and electric stoves Sampling inlet at face level in front of the cooker	SMPS TSI 3934; CPC TSI 3022A	Vegetable oil used to stir-fry 500 g of vegetables and also 5 rashers of bacon	No ventilation. All windows and doors were closed.
Wallace et al., 2004 USA	Domestic kitchen using gas stove Measurements performed in the duct of the ventilation system.	DMA Electrostatic classifier TSI 3071; CPC TSI 3010; APS TSI 3320; Optical particle counter model 500-I Climet Instruments	Deep frying (peanut oil) of flour tortillas; stir fry (peanut oil) vegetables and frying eggs with butter.	No ventilation. Forced ventilation (recirculation of air)

(a) DMA - Differential Mobility Analyser; SMPS - Scanning mobility particle sizer; APS - Aerodynamic particle sizer; CPC - Condenser Particle Counter

Table 5 Cont. Size distribution studies for cooking aerosols

Study and Country	Location and duration	Sampling method (a)	Food	Environmental condition
Wallace et al., 2006 USA	Personal and indoor (living room) measurements for 7 days in free-style living conditions.	Real time concentrations: Personal and indoor sampling using optical particle counter (personal MIE DataRAM) Integrated exposure: Personal – PEM gravimetric monitor Indoor – Harvard impactor monitor	Normal cooking activities	No control on ventilation
Hussein et al., 2006 Czech Republic	Domestic kitchen using an electrical stove and adjacent living room. Continuous measurement for 15 days at 3 min intervals Sampling ports at 1.5m from the ground and 1m (kitchen) and 5m (adjacent room) from the stove.	SMPS TSI 3934C;	Normal cooking activities (e.g. boiling potatoes, soup, rice, pasta, frying potatoes or pancakes, toasting and baking chicken in the oven.	Natural ventilation
See and Balasubramanian, 2006a, b Singapore	Domestic kitchen Inlets located 0.5 m above the gas stove	SMPS TSI 3034	Steaming, boiling, pan-frying, stir-frying, and deep-frying a pack of 150 g plain tofu (soybean curd) using corn oil.	No ventilation. All windows and doors were closed.
Sjaastad et al., 2008 Norway	Laboratory kitchen ($19m^2$) with electric stove in the middle of the floor with kitchen hood and adjoining room. Sampling ports 1m above the floor (all location) and 1.3 away from the stove (in the kitchen).	Kitchen: Particle counter Met One Model 237B; SMPS TSI-3936 Adjoining room : Electrostatic classifier TSI-3080 Ultrafine CPC TSI-2025A	Frying a beef steak with margarine at maximum power.	Mechanical ventilation.

(a) DMA - Differential Mobility Analyser; SMPS - Scanning mobility particle sizer; APS - Aerodynamic particle sizer; CPC - Condenser Particle Counter

Table 5 Cont. Size distribution studies for cooking aerosols

Study and Country	Location and duration	Sampling method (a)	Food	Environmental condition
Yeung and To, 2008 Hong Kong	Laboratory kitchen (168m^3) with gas stove and electric griddle. Fume hood installed above cooking area.	SMPS TSI 3734; Electrostatic classifier TSI 3071A; CPC TSI 3022A	Chinese style – frying vermicelli with beef- in gas stove; Western style – pan-frying steaks, chicken fillets or pork chops - in electric griddle and hot oil in electric griddle.	No ventilation
Buonanno et al., 2009 Buonanno et al., 2011 Italy	Open plan laboratory kitchen (80m^2) using gas and electrical stoves. Sampling 2 meters away from the stove for 8-10 mins.	SMPS TSI 3936 APS TSI 3321 CPC TSI 3775 Nanometer Aerosol sampler (TSI 3089)	Fry and grill different ingredients: pork meat, eggplant, chips and cheese, bacon and oils (olive oil, peanut oil and sunflower oil)	Minimum ventilation - doors and windows closed. Normal ventilation -doors and windows closed with mechanical ventilation in operation.
Buonanno et al., 2010 Italy	15 pizzerias Sampling 2 meters away from the stove for 8-10 mins.	SMPS TSI 3936 APS TSI 3321 CPC TSI 3775 Nanoparticle surface area monitor TSI 3550 ; PM_{10}, $PM_{2.5}$ and PM_1 measured using a DustTrak DRX Aerosol Monitor TSI 8534	Baking pizza	Normal commercial kitchen ventilation.
Glytsos et al., 2010 Czech Republic	Laboratory room (60m^3) Electric stove Sampling ports 0.9 m above the floor.	DustTrak Aerosol Monitor TSI 8520; P-Trak Ultrafine particle counter TSI 8525 ; GRIMM SMPS+C system - GRIMM, CPC Model 5.403 and Long Vienna DMA.	Frying half of an onion diced in hot olive oil (15 mL).	Mechanical ventilation using the air conditioning system.
Huboyo et al., 2011 Japan	In a kitchen (8.5 m^2) with fumehood and adjoining room (3 m^2). Cooking with single gas stove at medium setting. Sampling ports 1.1m away from the stove (in the kitchen) and 5 m away in the adjoining room.	Sioutas cascade Impactor (SKC); $PM_{2.5}$ UCB optical particle counter (Barkeley Air Monitoring Group	Frying in sunflower oil and boiling 400 g of soybean curd (tofu) and 400 g of chicken	Ventilation system (standard exhaust fan); Natural ventilation (windows opened)

(a) DMA - Differential Mobility Analyser; SMPS - Scanning mobility particle sizer; APS - Aerodynamic particle sizer; CPC - Condenser Particle Counter

Table 6 Particle diameter mode (i.e. diameter representing highest particle number concentration) of particle number size fraction distribution from cooking activities

Reference	Location	Comment	Diameter (nm)
Li et al., 1993	Taiwan	Frying Chicken	30-50
Siegmann and Sattler, 1996	Switzerland	Rapeseed Oil	30-100
Kleeman et al., 1999	USA	Meat charbroiling	180-320
Abt et al., 2000a,b	US	Size range Increasing diameter during cooking Oven cooking event	20-70
Wallace et al., 2004	USA	Cooking dinner	18-50
		Cooking breakfast	10-50
Yeung and To, 2008	Hong Kong	Frying vermicelli with beef	140
		Pan-frying steaks	150
		Pan-frying chicken fillets	115
		Pan –frying pork chops	102
		Hot oil test	107
Buonanno et al., 2009	Italy	Grilling in a gas stove at maximum power Bacon	50
		Cheese	40
		Eggplant	20
		Wurstel sausage	40
		50 g of chips fried with sunflower oil	50
		50 g of chips fried with olive Oil	61
		50 g of chips fried with peanut Oil	50
		50 g bacon grilled on a gas stove	60
Glytsos et al., 2010	Czech Republic	Frying a slice of onion with olive oil	20 – 45
Buonanno et al., 2011	Italy	Frying 100 g mozzarella	80
		Frying 100 g chips	60
		Grilling 100 g bacon	90
		Grilling 100 g eggplant	40

Table 7 Sampling, extraction and analysis of emission from cooking.

STUDY & RESEARCH OBJECTIVES	SAMPLING CONDITIONS	SAMPLE SUBSTRATE PRE-TREATMENT	EXTRACTION PROCEDURE	ANALYTICAL METHODOLOGY	COMPOUNDS ANALYSED
Rogge et al., 1991 Characterise organic compound composition emitted during meat charbroiling	1.8 µm cyclone upstream of 3 pumps Flow rate 9.0-9.6 L/min Sampling duration: 70-80 min	47mm teflon and quartz fiber	Samples were a composite of 15 quartz filters Extraction: Hexane (two times) and with benzene:2- propanol (2:l; three times) Extraction method: mild sonication Final volume reduced to 200-500 µL . Derivatization: one aliquot of the extract was derivatized with diazomethane to convert organic acids to their methyl ester analogues	GC/MS 30-m column	N-alkanes, branched alkanes, alkenes, alkynes, ketones,carbonyls,aromatic hydrocarbons, lactones, amides, saturated and unsaturated fatty acids, dicarboxylic acids, furans amides, steroids.
Wu et al., 1998 Determination of mutagenic PAH emitted from cooking oils	Personal sampling pump Flow rate: 2 l/min Sampling duration: 30 min.	37-mm Grade AA glass fiber filter paper	Extraction with a 200 ml acetone then concentrated to 10 ml in a vacuum rotary evaporator and evaporated to dryness under nitrogen stream. Residue was redissolved in 2ml for analysyis.	HPLC system (LH-20 column 15 mm id=190 mm) for PAHS. For detection of aminopyrenes a HPLC Hewlett Packard 1050 was used equipped with a 25-cm Nucleosil C column and spectrofluorimeter.	polycyclic aromatic hydrocarbons; nitro-polycyclic aromatic hydrocarbons
Schauer et al., 1999a Characterise organic compound composition emitted during meat charbroiling **Schauer et al., 2002** Characterise organic compound composition emitted during oil cooking	Emissions sampled in the ventilation system of a commercial kitchen downstream of the filter and grease extractor. Sampling time was 85 min. Dilution tunnel: mix exhaust emissions with 25- to 180-fold excess of HEPA filtered air. 1.8 µm AIHL-design cyclone separators upstream of samplers. Flow rate in each sampling train was 10 L/min, except sampling train a) at 30 L/min and sampling train g) at 0.2 L/min. Organic compounds collected using: a) 1 XAD coated denuder upstream of 3 quartz filters in parallel followed by 2 PUFs in series. b) 3 quartz filters followed each by 2 PUFs in series. EC/OC collected using: c) 2 quartz filters in series Mass emissions, trace metals and organic acids collected using: d) Teflon filter upstream of two KOH impregnated quartz fibre filters Mass emissions & soluble ions collected using: e) Teflon filter VOC collected using: f) 6-L SUMA canister downstream of teflon filter e) Carbonyls collected using: g) DNPH-impregnated C18 cartridges	Quartz fibre filters prebaked at 550ºC for 12 h Denuders coated following protocol described in Gundel et al. (1995) PUF plugs were pre-cleaned with 4 successive extractions of Dichloromethane/acetone/hexane (2:3:5). Solvent was removed by compressing the PUFs. Plugs were air dried in a dark organic free room, and stored in pre-cleaned glass jars at -20ºC.	*Quartz fibre filters:* Extraction: Hexane (two times) followed by benzene/2- propanol (2:l; three times) Extraction method: mild sonication *Denuders and PUFs:* Extraction: Dichloromethane/acetone/hexane (2:3:5) (4 times) Extraction method: Manual shaking In all cases, extracts from each aliquot were combined and concentrated to 250 µL Concentrated extract was split in two. Derivatisation: one aliquot of the extract was derivatised with diazomethane to convert organic acids to their methyl ester analogues. C18 cartridges were extracted as described in Grosjean et al. (1996) Teflon filters were extracted in water for water-soluble ions.	Organic compounds: Denuder, filter and PUF extracts (derivatized and underivatized aliquots) were analysed by GC/MS Hewlett Packard 5890 series fitted with a 30m, 0.25 mm inner diameter, 0.25 µm film thickness HP-1701 capillary column. Total non-methane organic gases and individual VOCs (C1-C10) were analysed from the SUMA canisters by GC/FID as described in Fraser et al. (1997). Carbonyl collected in the C18 cartridges were eluted with 2 mL acetonitrile analysed by LC/UV as described by Grosjean et al. (1996). Organic and elemental carbon (EC/OC) as described by Birch and Cary (1996). Trace metals were analysed by XRF. Soluble ions were analysed by AA and IC.	N-alkanes, branched alkanes, alkenes, alkynes, ketones, carbonyls, aromatic hydrocarbons, lactones, amides, saturated and unsaturated fatty acids, dicarboxylic acids.

Table 7. Cont. Sampling, extraction and analysis of organic emissions from cooking

STUDY & RESEARCH OBJECTIVES	SAMPLING CONDITIONS	SAMPLE SUBSTRATE PRE-TREATMENT	EXTRACTION PROCEDURE	ANALYTICAL METHODOLOGY	COMPOUNDS ANALYSED
Svendsen et al., 2002 Characterise aldehydes and fat aerosol collected in the breathing zone of the cook in fumes from commercial restaurants.	Personal exposure sampler with inlets located in the shoulder of the cook of 19 commercial kitchens using deep frying devices equipped with ventilation hoods. Aldehydes were collected a sampling device containing silica impregnated with 2,4-dinitrophenyl hydrazine. Flow rate was 1.5 L/min during 1.5-2.5 hours. Fat aerosol collected onto pre-weighted one glass fibre filter (Nucleopore AAA). Flow rate, 2 L/min during 65 to 200 mL. Total number concentration was measured with TSI 3936 SMPS used to measure the PAHs were collected onto glass fibre filters in a filter holder and 2 XAD-2 tubes downstream. Flow rate, 1 L/min during 200 min.	None	Fat aerosol extracted with 5 mL of 1,1,2-trichloro-1,2,2-trifluoroetahne. The aldehydes were reacted with 2,4-dinitropheynlhydrazin (DNPH) to form the corresponding stable hydrazone derivatives. The derivatives were eluted with HPLC grade acetonitrile.	Fat aerosol was determined using a FT-IR (Perkin Elmer 1605). The eluate was injected onto a C18 reverse phase column and detected using a UV detector operating at 360 nm.	Aldehydes, fat aerosol
McDonald et al., 2003 Characterise organic compound emission composition emitted during charbroiling and grilling of chicken and beef	University lab kitchen following commercial standard procedures. Emissions collected at the end of a residence chamber to allow the gas/particle equilibrium. 2.5 µm cyclone separators upstream of samplers. Flow rate in each sampling train was 113 L/min. Samples collected on Teflon filter for $PM_{2.5}$ and elements. Samples collected on quartz filters for carbon and ion analysis Samples collected on Teflon-impregnated glass fibre (TIGF) filter followed by a PUF/XAD-4/PUF sandwich cartridge for speciated particle-phase and semi-VOCs. CO was measured using non-dispersive infrared analyser.	Quartz fibre filters were baked at 900ºC for several hours. XAD-4 was solvent extracted in a Soxhlet with methanol followed by dichloromethane. TIGF filters were cleaned by sonication in CH_2Cl_2 for 30 min followed by another 30 min sonication in methanol. PUFs were rinsed with distilled water and Soxhlet extracted with hexane/ether (90:10), followed by acetone.	Half of the quart fibre filter was extracted with 10 mL of distilled-deionised water. TIGD filters and PUF/XAD-4/PUF sorbent were solvent extracted and combined for analysis.	$PM_{2.5}$ mass determined by gravimetric analysis. Ionic species determined by ionic chromatography. NH_4^+ was analysed by indolphenol automated colorimetry. Water-soluble K^+ was analysed by atomic absorption spectrometry. Carbon by thermal/optical reflectance. 0.56 cm^2 punch waa analysed for OC/EC by the TOR method. Elements by X-ray fluorescence. Organics determined with an Agilent GCMS (GC Model 6890plus, MSD Model 5973N) equipped with a 60m x 0.25mm x 0.25 um DB5-MS capillary column.	$PM_{2.5}$, CO, OC/EC, inorganic species, elements, lactones, sterols, PAHs, biphenyls,
Zhu and Wang, 2003 Characterise PAH emitted in commercial and domestic Chinese kitchens	A sampler was located in a new kitchen 0.5 m from the pan (cooking methods) and in the centre of the kitchen (domestic and commercial kitchens). In all cases, the sampler was 1.5 m above the ground level. All doors and windows were closed during cooking. Electric hobs were used for cooking. Samples were collected over 100 mins to test different cooking methods, and over 2 consecutive days for 12-h (8:00 – 20:00) in domestic and commercial kitchens. Low noise small samplers (MP-15CF) operated at 1.0 l/min equipped with a Whatman glass filter (GFF, 25 mm) collected particle bound PAHs and a XAD-2.5 g cartridge collected the gaseous PAHs.	Filters were combusted overnight and sealed in aluminium foil. XAD-2 cartridges were pre-extracted in dichloromethane and methanol for 48 h, vacuum-dried in desiccators and stored in solvent rinsed glass jars.	Extraction by sonication for 30-min with a 20 ml mixture of DCM and acetonitrile (3:2). The extract was concentrated to 10 ml and 30 µl of dimethyl sulfoxide was added. The mixture was concentrated under nitrogen and 1ml of methanol was added. 100 µl were injected for analysis.	HPLC (Hitachi L-7000 series) consisting of a precolumn (Supelco, 5C-18, 4.6x 50 mm) for PAH condensation and cleanup, a main column (Wakosoil, 5C-18, U4:6 250 mm) for separation and a fluorescence detector.	PAHs

Table 7 Cont. Sampling, extraction and analysis of organic emissions from cooking

STUDY & RESEARCH OBJECTIVES	SAMPLING CONDITIONS	SAMPLE SUBSTRATE PRE-TREATMENT	EXTRACTION PROCEDURE	ANALYTICAL METHODOLOGY	COMPOUNDS ANALYSED
Chen and Chen, 2003 Characterise PAHs in fumes during frying of chicken.	Emissions collected on adsorption wool fitted on the cover of frying tank (closely tight during sampling)	Adsorption wool	Soxhlet extraction for 20hrs using acetone to 1ml then evaporated to dryness then residue dissolved in 10 ml acetone and stored for GCMS analysis.	GC/MS equipped with an HP-5MS (30 m x 0.25 mm i.d., 0.25 um film thickness)	PAHs
Li et al., 2003 Characterise PAHs in fumes during cooking of different styles	Emissions collected isokinetically from the exhaust vent in commercial kitchens. Three consecutive samples were collected at 10L/min for 45 min during the cooking time. Particle bound PAHs were collected on a tube-type glass fibre thimble (25x90 mm). Gaseous PAHs were collected onto a 5-cm polyurethane foam (PUF) followed by a 2.5 cm Xad-16 resin cartridge supported by a 2.5 cm PUF.	Samples were kept prior and after sampling in cleaned screw-capped glass bottles and jars.	Samples were extracted in a Soxhlet extractor with 1L of mixed solvent n-hexane/dichloromethane (1:1) for 24 hours. The extract was concentrated, cleaned and re-concentrated to 1 or 1.5 mL.	Hewlet-Packard GC HP 5890A with a Mass Spectrometer dector HP 59H72 equipped with a HP Ultra 2 50m x 0.32 mm x 0.17 um column.	PAHs
He et al., 2004b Characterise fumes emitted during Chinese style cooking	Samples collected at the exit of the exhaust vent of two commercial kitchens. Sampling times were 90-120min at lunchtime and dinner. Samples collected onto two honeycomb sampler and a three stage cascade impactor to collect $PM_{2.5}$ at 25 L/min. One honeycomb contained PTFE filters for particle mass determination and and ionic species analysis. The second honeycomb and the cascade impactor were loaded with quartz filters (Pallflex 2500QAT-UP) for the determination of EC/OC and organic speciation.	Quartz fiber filters were baked for 4 hours at 500ºC. Pre- and post-sampling filters were stored in pre-cleaned 250 mL glass jars with 3-5 mL of methylene chloride to prevent microbial growth. Sampled filters stored in the freezer.	Samples extracted with dichloromethane (3 times) and methanol (2 times) for 20 min using a mild ultrasound bath. Reduced to 5 mL with rotary evaporation and further concentrated to 1ml under a N_2 stream and split into three fractions. Two fractions were derivatised with BF_3/CH_3OH and bis-(trimethylsilyl) trifluoroacetamide (BSTFA) plus 1% trimethylchlorosilane (TMCS) to convert organic acids and unmethylated compounds to their methyl ester and trimethylsilyl derivatives respectively. Derivatisation temperatures and times were 80oC for 30 min and 85oC for 40 min respectively.	$PM_{2.5}$ mass determined by gravimetric analysis. Ionic species determined by ionic chromatography (DX-600, Dionex Corp). EC/OC determined with the Sunset analyser. Organics determined with an Agilent GCMS (GC Model 6890plus, MSD Model 5973N) equipped with a 60m x 0.25mm x 0.25 um DB5-MS capillary column.	N-alkanes, n-fatty acids and dicarboxilic acids; PAHs and other compounds including cholesterol and levoglucosan.
He et al., 2004c Characterise fumes emitted during Chinese style cooking	Samples collected at 40-60 cm at leeway from the exhaust vent of two commercial kitchens. Sampling times were 100-120min at lunchtime, and 45 minutes at dinner. Samples collected onto quartz fibre filters with a three stage cascade impactor (<10um, 10-2.5 um and <2.5 um) at 25 L/min.	Quartz fiber filters were baked for 2 hours at 500ºC. Pre- and post-sampling filters were stored in pre-cleaned 250 mL glass jars with 3-5 mL of methylene chloride to prevent microbial growth. Sampled filters were stored in the freezer.	Samples extracted with methylene chloride (3 times) for 20 min using a mild ultrasound bath. Reduced to 5 mL with rotary evaporation and further concentrated to 1ml under a N_2 stream.	GC/MS Autosystem XL Gas Chromatography coupled with a TurboMass Mass spectrometry (Perkin Elmer) equipped with a 60m x 0.32mm x 0.25 um fused silica capillary column (PE-35MS)	N-alkanes, n-alkenes, n-fatty acids; n-alkanal; n-alkenals; PAHs

Table 7. Cont. Sampling, extraction and analysis of organic emissions from cooking

STUDY & RESEARCH OBJECTIVES	SAMPLING CONDITIONS	SAMPLE SUBSTRATE PRE-TREATMENT	EXTRACTION PROCEDURE	ANALYTICAL METHODOLOGY	COMPOUNDS ANALYSED
See et al., 2006; See and Balasubrabramanian, 2006b Characterise PAH and metal composition emitted during Chinese, Malay and Indian style commercial cooking **See and Balasubrabramanian, 2006a, 2008** Characterise emissions from 5 types of cooking methods (steaming, boiling, stir-frying, pan-frying and deep- frying)	Sample collected at 1.5m above ground level at the opposite site of a 4 LPG burners stove in commercial food stalls.(See et al., 2006; See and Balasubrabramanian, 2006b) Sample collected at 1.5m above ground level and 0.2 m from a 2-burner domestic stove with no ventilation. Samples collected during cooking activities. (See and Balasubrabramanian, 2006a, 2008). Samples collected for 12 hours during cooking and non-cooking activities. A MiniVol portable air sampler (Airmetrics) collected $PM_{2.5}$ at a flow rate of 5 L/min onto: - 47mm 2 µm PTFE Teflon filter for gravimetric, metals and ion analysis. - 47mm QMA quartz filters for PAH	QMA filter was pre-combusted at 400ºC for 24h prior to sampling. No pre-treatment of Teflon filter	*PAH* Microwave extraction using 20mL acetone:hexane (1:1) for 20 min at 150W. Extracts concentrated to 3 mL using a rotary evaporator. Further concentration to almost dryness with N_2 stream and reconstituted with 1 mL of extraction solvent. *Metals* Microwave extraction as described by Swami et al. (2001) *EC/OC* 2 6mm punches of a quartz fibre filter. One punch was combusted at 350ºC for 24h to remove the OC.	*PAHs* Hewlett Packard 6890 series GC/MS fitted with a DB-5MS 5%-phenyl-methylpolysiloxane 30m x 0.2 mm internal diameter x 0.25 µm film thickness. *Metals* Perkin Elmer ELAN 6100 ICP/MS *EC/OC:* Both combusted and uncombusted punches were analysed for carbon using a 2400 Series II CHNS/O analyser (Perkin Elmer) operated at the CHN mode with acetanilide as calibration standard.	PAHs Metals EC/OC
Zhao et al., 2007 b, c Characterise organic compound emission composition emitted during Chinese and Western style cooking	Emissions sampled at the exhaust vent of the ventilation system of commercial kitchens downstream of the filter treatment methods. Samples collected during rush hour at lunch and dinner times. Sampling time was 120 min. 2 medium-volume samplers at a flow rate of 78 L/min collected samples in 90mm quartz fibre filter. 2 Dustraks (TSI) monitored the relative concentrations of $PM_{2.5}$ and PM_{10}. Background $PM_{2.5}$ was collected in the city using a hi-volume sampler (Andersen).	Quartz fiber filters were baked at 450ºC for 4.5 hours. Prior to sampling, filters were stored in a freezer.	Extraction with three successive portions of dichoromethane and methanol (3:1) for three 15-min in the ultrasound bath at room temperature. Extracts were filtered and destilled under negative pressure to 3-5 mL, subsequently concentrated to 1 mL under N_2 gas stream, and divided in three portions: Portion 1 – analysed directly in GCMS for non polar organic compounds. Portion 2 - Derivatized with BSTFA plus 1% TMCS at 70ºC for 2 h. This was analysed for polar organic compounds. Third portion – stored at 4oC as a backup.	Organics Agilent 6890plus / MSD model 5973N GC/MS using a DB-5MS 60m x 0.25 mm internal diameter x 0.25 µm film thickness column EC/OC: Carbon analyser Sunset Lab.	N-alkanes, PAHs, N-alkanals, N-alkanones, lactones, amides, saturated and unsaturated fatty acids, dicarboxylic acids, anhydrides, sterols

Sjaastad and Svendsen, 2008; Sjaastad et al., 2010; Sjaastad and Svendsen, 2009 Characterise PAHs, aldehydes and particulate matter collected in the breathing zone of the cook in fumes from frying a beefsteak.	Model kitchen (19 m2) containing gas or electric hobs and a canopy fume hood. Personal exposure sampler with inlets located in the shoulder of the cook. PAHs were collected onto glass fibre filters in a filter holder and 2 XAD-2 tubes downstream. Flow rate, 1 L/min during 200 min. Aldehydes were collected into stainless steel sorbent tubes filled with 220 mg Tenax TA. Flow rate, 100 mL/min for 10-200 min. Total particles collected onto pre-weighted double Gelman glass fibre filters. Flow rate, 2 L/min during 65 to 200 mL. Total number concentration was measured with TSI 3936 SMPS.	None	PAH were desorbed in dichloromethane.	PAH measured by a commercial laboratory following a modified version of AMI L5, NIOSH 5515, ISO/CD 12884 and VDI 3873. Aldehydes measured using an automatic thermic desorption unit ATD 400 (Perkin Elmer) connected to a GCMS (Focus GC-DSQ, Thermo Electron Corporation).	PAH; aldehydes

Table 8 Main identified cooking marker species in the literature

COMPOUND ANALYSED	SOURCE IN FOOD	OTHER SOURCES IN THE ENVIRONMENT
Unsaturated fatty acids Oleic acid- 9-octadecenoic acid-meat tracer, canola oil (Schauer et al, 2002) Linoleic acid- 9,12-octadecadienoic acid Palmitoleic acid- 9-hexadecenoic acid meat cooking (Zhao, 2007a; Robinson et al, 2006)	Combustion of triglycerides and phospholipids from seed oil, vegetable oil, fats of animals and meet cooking (Robinson et al, 2006)	Biomass smoke, motor vehicle exhaust and road dust (Robinson et al., 2006)
Saturated Fatty Acids hexanoic acid octanoic acid nonanoic acid-from seed oil (Schauer et al, 2002) hexadecanoic acid, palmitic acid, (Robinson et al, 2006)	Combustion of triglycerides and phospholipids from seed oil, vegetable oil and fats of animals. The acids are formed directly from the pyrolysis of their glycerol ester precursor analogues (nonanoic acid formed from the breakdown of oleic acid present in seed oil (Schauer et al, 2002)	Palmitic acid are emitted also from biomass smoke, motor vehicle exhaust, road tire dust (Robinson et al, 2006), tyre dust cigarette smoke, roofing asphalts and fuel combustion (Nolte et al., 1999)
Dicarboxylic Acids-C4-C8 hexanedioic acid – from meat cooking and seed oil octanedioic acid – from seed oil nonanedioic acid, tetradecanoic acid, octadecadienoic acid from soybeans oil (Schauer et al, 2002)	Products of dialdehydes formed during auto oxidation of unsaturated lipids. Produced from meat cooking (C_4-C_8 high concentrations for hexaneoic acid) and heating of seed oil (C_8 higher concentrations) (Zhao, 2007a).	
Polycyclic Aromatic Hydrocarbons pyrene chrysene –seed oil and meat cooking (Zhao, 2007a) benzo[a]pyrene	Incomplete combustion of organic substance (cooking materials such as meat, vegetables, oil)	House heating Cigarette smoking (Kleeman et al., 2008). Heavier PAH (coronene, benzo[ghi]perylene, indeno[1,2,3-cd]pyrene) are emitted from motor vehicles and retene from biomass burning (Brinkman et al., 2009)
Molecular bio markers • **Monosaccharide Anhydrides-** from breakdown of cellulose during cooking (Zhao, 2007a) Galactosan Mannosan levoglucosan • **Sterols** □-sitosterol –present in animal and vegetable body tissue (Zhao, 2007a). Cholesterol – from meat cooking (Zhao, 2007a; Robinson et al, 2006) stigmasterol	From the organic compounds of biological origin which have restricted occurrence and molecular stability so can be detected in body tissues. Plant lipid membranes and waxes. For Chinese food the average ratio of levoglucosan/(mannosan+galactosan) is 12 (Zhao, 2007a).	Levoglucosan is released from wood burning. (Kleeman et al., 2008; Zhao et al., 2007a Cholesterol produced from cigarette, debris of plant and road dust (Zhao et al., 2007a; Robinson et al., 2006; Nolte et al., 1999).
N Alkanes C_{23}-C_{31} C_{23}-C_{31} from cooking material/contents (Zhao, 2007a)	From cooking material/contents (Zhao, 2007a)	From motor vehicles (Brinkman et al., 2009)
Lactones C_7-C_{18} From food cooking (Zhao, 2007a; Schauer et al., 2002). 5-propyldihydro-2(3H) furanone (Schauer et al, 2002) 5-dodecyldihydro-2(3H) furanone (Schauer et al, 2002)	Meat charbroiling and food cooking (Schauer et al, 2002)	
Alkanals and alkanones C_9-C_{15} from cooking oil Nonanal (Zhao, 2007a) 2-pentadecanone- from soybean oil and seed oil (Schauer et al, 2002) 2-nonanone 2-undecanone 2-pentadecanone	Combustion of triglycerides in oil (Zhao, 2007a). From the decomposition of unsaturated fatty acids (oleic acid) (Schauer et al, 2002)	
Inorganic elements and ions From meat cooking Aluminium (Schauer et al, 1999a) Silicon (Schauer et al, 1999a) Phosphorus (Schauer et al, 1999a) Sulphur (Schauer et al, 2002; Schauer et al, 1999a) Chlorine (Schauer et al, 1999a) Potassium (Schauer et al, 1999a) Sodium (Schauer et al, 2002; Schauer et al, 1999a) Nitrate (Schauer et al, 2002; Schauer et al, 1999a)	From meat, vegetables and sauces (Schauer et al, 2002)	From soil, motor vehicles, cigarettes and biomass burning (Brinkman et al., 2009)

1.8 Effect of cooking styles and ingredients on organic compound emission profiles

Research has identified that different cooking styles emit different profiles of compounds. The differences have been attributed to factors such as cooking processes and ingredients (Hildemann et al., 1991a, Rogge et al., 1997, Schauer et al., 1999a, He et al., 2004a). Western fast food cooking involves frying with beef and chicken as the main cooking method and meats consumed. Chinese cooking practice on the other hand generally involves the use of pork, poultry, seafood as well as vegetables during cooking as listed in Table 2. Chemical composition variations are thus expected to be observed for various different cooking operations. For instance, nonanedioic acid has been identified as the most abundant dicarboxylic acid in Chinese cooking and hexanedioic acid for meat cooking (He et al., 2004d, Rogge et al., 1991, Zhao et al., 2007b). Sitosterol and monosaccaride anhydrides have been attributed to vegetables used in Chinese cooking as they were not observed in meat cooking processes. These differences in chemical composition need to be considered for selection of molecular markers, which will be useful to assess the contribution of cooking to atmospheric particulate organic matter (POM) (Rogge et al., 1991, He et al., 2004c, Zhao et al., 2007b, Zhao et al., 2007a). Figure 2 shows Marker-to-OC ratios of meat cooking profiles using profiles from Rogge at al. (1991), Watson et al. (1998) and Schauer et al. (1999a; Schauer et al., 2002). These source profiles and species are usually included in models by normalising emissions with OC or $PM_{2.5}$.

Higher fat contents in cooking ingredients have been found to produce more fatty acids compared with the low fat content ingredients in the same cooking operation (Zhao et al., 2007a, Zhao et al., 2007b, Rogge et al., 1991). This is generally observed also when Chinese cooking is compared with Western style fast food; the latter having higher concentrations of fatty acids, indicating the high proportion of ingredients with higher fat content. Animal and

vegetable fats are rich in high concentrations of normal fatty acids with even carbon numbers from C4 to C34 as triglycerides and phospholipids (Zhao et al., 2007a).

In a experiment comparing grilling and charbroiling different types of meat, grilling was found to emit less organic compounds than charbroiling, which yielded about 5 times more PAH (i.e. 30-50 mg/kg for charbroiling vs. <10 mg/kg for grilling), 10 times more lactones (i.e. 7-30 mg/kg for charbroiling vs. 2-4 mg/kg for grilling) and 20 times more cholesterol (i.e. 1-8 mg/kg for charbroiling vs. 0.04-0.2 mg/kg for grilling) (McDonald et al., 2003) .

When different types of meat were grilled in a shed, Mohr et al. (2009) reported large differences of emissions with increasing emissions as the fat content increased, even when the meats were cooked in the same manner. This is qualitatively consistent with earlier studies (McDonald et al., 2003, Rogge et al., 1991). Rogge et al. (1991) reported that generally grilling of meat led to the higher production of aerosols made of fatty acids. This was attributed to the oil and grease droplets falling into the gas flame or onto the heat source where they would vaporize and renucleate and grow into small particles.

Zhao et al. (2007a) investigated the chemical composition of particulate organic matter from Western fast food cooking and identified tetradecanoic acid, hexadecanoic acid, octadecanoic acid, 9-octadecanoic acid, nonanal, levoglucosan, hexanedioic acid and nonanedioic acid as potential tracers with saturated and unsaturated fatty acids accounting for 78% of total quantified compounds. When they analysed the chemical composition of aerosol from Chinese cooking, they identified also a dominant homologue of fatty acids with its concentration being about 73-85% of the quantified compounds. They also identified levoglucosan and β-sitosterol as well as a clear pattern of n-alkanes which were taken as an indication of vegetables consumed in the Chinese cooking process (Zhao, 2007b). The concentration of quantified compounds per unit of particulate organic matter in Western cooking was found to be much higher than that in Chinese cooking (Zhao, 2007a). The candidate organic tracers that they

found to distinguish emissions of Western cooking from Chinese cooking in Ghanzou (China) are tetradecanoic acid, hexadecanoic acid, octadecanoic acid, oleic acid, levoglucosan, mannosan, galactosan, nonanal and lactones (Zhao, 2007b). Table 9 shows clearly from their findings that the Chinese cooking made a much greater contribution of PAHs to particulate organic matter than Western fast food with 2,855 ng/mg of particulate organic matter in Chinese cooking as against 40 ng/mg in Western cooking.

Nolte et al. (1999) analysed meat cooking smoke and found that 1-palmitin and 2-palmitin were the most abundant compounds observed with significant emissions of 1-stearin and 1-olein monoglycerides and cholesterol (Nolte et al., 1999). Similar to what was observed with emissions of particulate number and particulate matter mass, higher concentrations of organic pollutants were observed to be emitted during oil-based cooking methods compared to steaming and boiling which were water-based (See and Balasubramanian, 2008). Also an analysis of commonly used cooking fuels in Hong Kong identified that gas cooking produced higher concentrations of PM_{10}, organic material and total volatile organic compounds during cooking by stir frying, pan frying and deep frying in domestic settings (To and Yeung, 2011). Higher concentrations of pollutants were observed in commercial kitchens compared to domestic kitchens probably due to the volume of food cooked or methods of cooking used. In the commercial restaurant, broiling of meat was found to produce higher concentrations of PM and VOC especially for electric broiling of meat compared to gas broiling. This was attributed to a larger contact area for the beef on the electric broiler compared to the gas broiler leading to more intense effect of the heat (To and Yeung, 2011).

An analysis of occupational exposure to cooking fumes was carried in a laboratory kitchen to investigate exposure of cooks to polycyclic aromatic hydrocarbons (PAHs), higher mutagenic aldehydes, total particles, and ultrafine particles during cooking (Jorgensen et al., 2013). The level of total particles was between 2.2 and 4.2 mg/m3 with statistically significant higher amount of ultrafine particles generated when a gas stove was used compared with frying on an electric stove. The gas flame was also observed to release particles but not enough to contribute

to the difference observed between the two fuel types. The amount of total PAH observed was of 270–300 ng/m3 air when fresh bacon pan fried with a high concentration of retene observed to when smoked bacon was fried. Another laboratory experiment by Sjaastad, et al, in 2010 where they looked at exposure to PAH, mutagenic aldehydes, and particles during the frying of beefsteak on a gas stove and on an electric stove using different types of vegetable frying fat showed somewhat higher levels of naphthalene which was the only PAH found in all the samples in the study by Jorgnsen et al,2013.

In a recent study analysing frying found that deep-frying generated more PAHs and benzo[a]pyrene (B[a]P) (1.3 and 10.9 times, respectively) than any other frying method and this was attributed to the volumes of edible oil used in deep frying and also the high oil temperatures relative to other frying methods (Yao et al., 2015). Total B[a]P concentration of deep-frying was found to be 2.2-fold larger than that of regular frying with rapeseed oil producing high PAH emission than soybean, peanut, and olive oil.

Table 9 Concentrations of organic compounds from western-style fast food and from Chinese cooking (ng/mg of particulate organic matter) (Zhao et al., 2007b,c)

Organic compounds	Western-style fast food cooking	Chinese cooking
n-Alkanes	3860	1880
Polycyclic aromatic hydrocarbons	40	2860
n-Alkanones	22700	2440
n-Alkanals	29200	3440
Lactones	13300	2140
Amides	4690	531
Saturated fatty acids	374700	26800
Unsaturated fatty acids	93300	29030
Dicarboxylic acids	57900	2050

Monosaccharide anhydrides	97	314
Sterols	487	1680

1.9 Research aims and objectives.

Cooking has been identified as a source of primary organic aerosol but it is still not a thoroughly understood source of primary organic aerosol. Generally a scarcity of literature has been identified on cooking, with the few studies available being from different geographical locations and restricted to only the cooking styles of the selected locations. Not much work has been carried out analysing emissions from different styles and methods of cooking during the same study period. This study will attempt to address these gap.

The aim of the research is to assess the emission profiles from various culinary techniques and also to use the emission profile for source apportionment.

Specific objectives are:

1. Examine the chemical composition of particulate organic matter in particulate matter emitted during different cooking styles.
2. Identify and characterise organic tracer species from different cooking styles that will be useful in factor analysis for source apportionment (source profile).
3. Use personal monitoring of people while cooking in their kitchens using a range of ingredients to assess exposures.
4. Apply source profile developed in source apportionment to determine and quantify contribution of cooking to particulate matter in a built up area.

This thesis will analyse particulate matter emission from cooking using various types of cooking methods- Chinese, Indian, western and African styles.

1.10 Hypothesis

Chinese cooking style is associated with a higher quantity of emission of organic compounds as compared to Indian, African and Western style of cooking.

1.11 Data Analysis

Microsoft Excel and SPSS 21.0 were used for data analysis. Receptor modelling was performed using the USEPA CMB Model 8.2. Details of the CMB model are described elsewhere (chapter 5).

1.12 Thesis organisation and structure

The thesis chapters shall be organized as follows:

Chapter 2- Methodology -Sampling design and chemical analysis.

Chapter 3- Source profile-Trailer sampling.

Chapter 4- Ambient samples- Real kitchen

Chapter 5- CMB- Using profile obtained as input to analyse existing data to apportion contribution from cooking

Chapter 6- Conclusion

Chapter 7- Recommendation and future directions.

CHAPTER 2 -Methodology -Sampling design and chemical analysis.

Overview

This chapter highlights sampling methods, locations and chemical analysis carried out on samples collected.

2.1 Sampling locations

2.1.1 Trailer kitchen design-

2.1.1.1 Overview

Different culinary techniques listed in Table 10 were used to cook chicken in the laboratory trailer kitchen and PM samples for cooking-related emissions were collected on 47 mm filters (Teflon and quartz fibre) from the exhaust of a designed kitchen. In this kitchen no interference exist form any other activities. Chicken was used as the standard ingredient in order to reduce variation between various cooking styles. The style of cooking was selected based on existing literature of studies that have been carried out on cooking experiments listed in chapter 1 (there is a good volume of a few studies that have been done on Chinese, Western and Indian cooking but none of African style of cooking) . Another factor for choosing style of cooking was because of the diverse population of Birmingham,, so randomly cooking styles were selected

The design of the trailer kitchen consisted of a 70 cm baumatic chimney hood with an extraction capacity of 500 m^3/hr. with no grease filters or baffles in the hood the exhaust flow was measured using an anemometer AVM 07 to determine and confirm the flow rate of the extractor fan. The anemometer was placed at the face of the exit of the duct pipe to determine the velocity of air from the extractor which was multiplied with the area of the duct.

3 anemometer readings were taken and the average value of the velocity was 12.5m/s.

Area of the duct- width x height (from Table 11) = 0.011m^2

Therefore velocity of air extracted by the hood=12.5 x 0.011=0.1375m^3/sec=495m^3/hr.

The flow rate was found unaffected and lie within 495 and 500m^3/hr without the filters/ baffles.

The work table height (counter on which cooker would be placed) table was 870 mm from the floor and 40mm wide for the gas and electric hob to be placed on it. Depending on what heat source was to be used on a particular sampling day (electric hobs or gas hob) which was placed on the work table constructed with an average thickness height of 40 mm making total height from ground to cooking surface 910mm. The work table was constructed with dimensions 870 mm height by 600 mm by 680 mm. The distance from fume source to face of hood was about 61cm from the cooking source.

The particles were collected on quartz fibre filters directly from downstream of the extractor pipe system of the ventilation system above the heat source where cooking exercises was taking place for sampling. The sampling times were between 45–90 min per sample with 6 samples taken for every cooking style. Pump of 30 L/min was used attached to filters.

Table 10 Cooking styles and food option selected.

Cooking style	**Dish**	**Method**
Chinese	Chicken kun pao with rice	Stir fry
Western	Chicken, eggs and chips	Deep fry
Indian	Chicken tikka masala with rice	Stew
African	Chicken in tomatoes stew with rice and plantain	Deep frying, stew

2.1.1.2 Dimension for sampling setup

To ensure a laminar flow during sampling, it is important to determine the point to sample from along the extractor exhaust duct.

Table 11 Dimension of extractor duct

Length(l)	Width (w)	Height (h)	l*w(m^2)	w*h(m^2)	h*l(m^2)	total(m^2)	total surface area (m^2)
1.7	0.2	0.055	0.34	0.011	0.0935	0.4445	0.889

Total surface area of duct -a=2((l&w)+(w*h)+(l*h))

area cross section=width*height=0.011

flow rate= velocity *area……(flow rate for hood -500m^3/hr)

therefore 500m^3/hr= velocity*0.011

vel=45454.55m/hr

Sampling pump

flow rate=30l/min=1.8m^3/hr

area(area of sampling pipe)=flow rate/ velocity of hood

area=1. 8m^3/hr *45454.55m/hr=3.96E-05m^2

area of a circle (cross sect area) is=πr^2 (r=radius of pipe)

0.0000396= πr^2

r= 0.003549003m=> diameter =2*r=0.007m=0.7cm=**7mm**

Location of Sampling Port

"*To ensure laminar flow, sampling ports shall be located at least 8 times chimney diameter down stream and 2 times up stream from any flow disturbance*"(CPCB, 2007).

The equivalent diameter (De) for a rectangular cross section, such as the hood, shall be calculated by using following equation to determine location and distances to place sampling port.

$$De = \frac{2LW}{L + W}$$

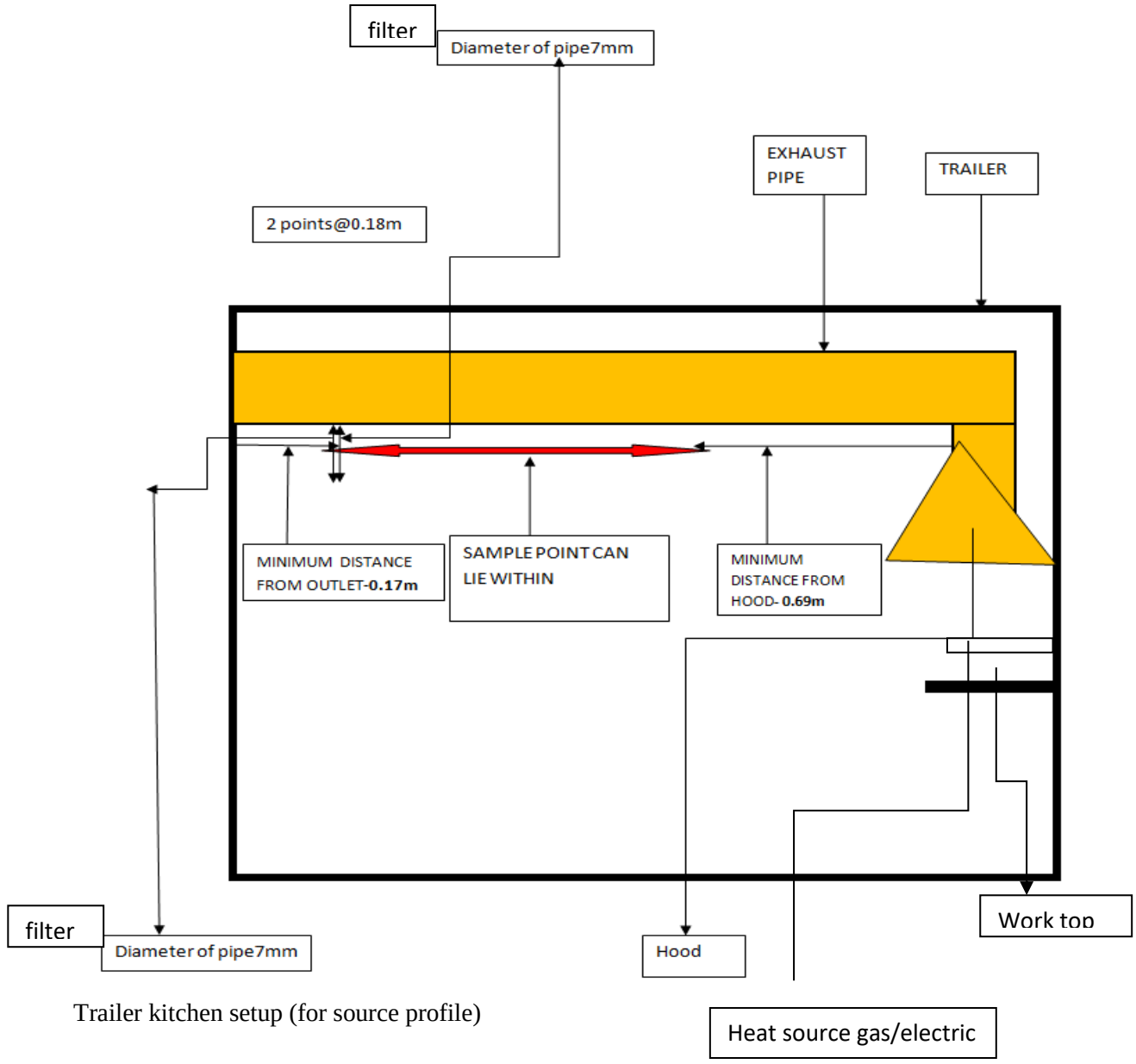
Where L =Length in m, W= width in m.

From table De= 0.086275m

Sampling point=minimunm 8*equivalent diameter=0.0863*8=0.69m=69cm from source

From other side (outside)=2* equivalent diameter =0.0863*2=0.1726=17cm from the vent.

Diagram of the kitchen setup is shown in **Figure 5**. **Figure 6** and **Figure 7** are pictures taken in the trailer showing the setup.



Trailer kitchen setup (for source profile)

Figure 5 Trailer setup

2.1.1.3 Ingredients and recipe for cooking

RECIPES.

Below are details of steps taken to cook the various foods.

Table 12 Cooking style and ingredients

S/N	COOKING STYLE	Method	INGREDIENTS	Oil	spices	DURATION (Minutes)
1	CHINESE (kung pao chicken)	Stir fry	Meat -chicken Vegetable- green bell peppers, celery, Chinese cabbage, water chestnuts, and carrots OTHER-roasted peanuts,chopped, sliced, or diced red bell peppers, rice and chili peppers.	Peanut oil	essence of chicken, salt, peanut oil, light soy sauce	45
2	WESTERN (Chicken, eggs and chips)	Deep fry	Meat- chicken Other- potatoes , eggs	Sun-flower oil	Salt	30
3	INDIAN FOOD (chicken tikka masala)	Stew	Meat type- chicken, Other –Rice, lemon juice, yogurt, garlic, tomatoes, chilli, cream.	Mustard oil	Cardamom, turmeric, garam masala, cinn amon	60
4	AFRICAN (Nigerian chicken stew and plaintain)	Deep frying, stew	Meat- chicken Others- rice, plantain, chilli, onion	Ground nut oil	Thyme, curry	60

Quantity of ingredients used in the study.

STYLE	**AFRICAN**	**CHINESE**	**WESTERN**	**INDIAN**
INGREDIENTS	Chicken-1kg, Onion-100g, Rice-400g, Tomatoes-1.4kg, Plaintain-500g, Chili-100g, Ginger-5g, Bell pepper-150g	Chicken-1kg, Onion-100g, Rice-400g, Tomatoes-1.4kg, Stir fry vegetables-350g, Peanuts-20g, Ginger 5g	Chicken-1kg, eggs-4pieces, potatoes-600g	Chicken-1kg, Onion-100g, Rice-400g, Tomatoes-1.4kg, Ginger-5g, Yogurt-150g, Cardomin pods-8pieces, Bell pepper-150g, ginger 5g.

HEAT SETTING USED FOR ALL COOKING –180^{O}C and later 200^{0}C

Indian chicken masala

http://www.bbc.co.uk/food/recipes/chickentikkamasala_67780.pdf

1. The chicken was cut into bite-size pieces and mixed with tandoori paste and yoghurt and left marinate in a non-metallic dish for some time while stirring occasionally.
2. Oil was heated in a deep frying pan, and when very hot cinnamon, cardamom pods and onion were added. These were fried for about 5-6 minutes until they began to brown, then ginger, garlic, cumin, coriander, turmeric and cayenne pepper were added.
3. After the spices were cooked for about a minute the chicken and marinade were added and fried for 3-4 minutes, then tinned tomatoes, chicken stock or water, garam masala, lemon juice and salt were added. The curry was then left to a simmer and cook on a low heat for about 30 minutes.
4. Rice was boiled after the curry was ready.

Chinese Kung pao

(http://www.tastebook.com/recipes/1944936-Kung-Pao-Chicken):

1. The chicken meat were cut into small cubes and rinsed in water and marinated with the ingredients above for 30 minutes.
2. The sauce ingredients were then mixed in a small bowl and set aside.
3. The wok was heated up with one tablespoon cooking oil in it and the marinated chicken was stir-fried until they were 70% cook. When fried the chicken was set aside.
4. The wok was cleaned and 2 tablespoons of cooking oil was added into it until it smoked.
5. Ginger and garlic slices were added to the wok and stir fried then dried red chillies were added.
6. The dried red chillies were stir fried until aromatic and smell spicy then the chicken meat was added.
7. Roasted peanuts were then added and stir frying continued.

8. The sauce was then added and stirred continuously until the chicken meat was coated with the sauce.
9. The scallions were then added and stirred evenly.
10. Rice was boiled.

Nigerian chicken stew

http://www.ihiagwa.com/cusines.htm

1. The chicken was washed and cut into 10-12 pieces
2. The chicken was seasoned with salt add the sliced onions thyme curry and cooked for 30-40 minutes until tender.
3. Oil was heated up in a pan and the chicken was fried until brown but not too dry. In another pot, oil was heated up and ground onions, chillies and tomatoes were heated for 20minutes until fairly dry.
4. Tomato puree was added to the oil. This was stirred thoroughly and later the fried chicken pieces were added.
5. The tomatoes were allowed to cook and simmer gently for another 10 minutes while stirring frequently.
6. Excessive oil that rises to the top were drained off.
7. Rice water and plaintains were fried in oil.

Figure 6 Frying plaintain in trailer kitchen

Figure 7 Trailer kitchen setup

Sample size was obtained using the DSS RESEARCH TOOL software online, https://www.dssresearch.com/knowledgecenter/toolkitcalculators/samplesizecalculators.aspx.

Concentrations of compounds obtained from previous studies are used to calculate the minimum sample size. For instance based on previous studies of cooking (He et al, 2004; See et al, 2006; Zhao et al 2007), we expect an average concentration of 278ng/mg of POM and a standard deviation of 68 for nonacosane for cooking Western style and an average

concentration of 258ng/mg of POM and a standard deviation of 60 for the same compound for cooking Chinese style. The sample size calculation considering an independent t-test required 2 samples to detect any significant differences at a confidence level of 0.05 and a statistical power of 90%. Our suggested sample size (6 samples) hence would allow to detect differences between these two different cooking styles. This was carried out for several other compounds and between 1 and 2 samples were the number obtained.

Summary of samples from trailer-

Trailer sampling- 6 for each cooking style-(one Teflon and quartz fibre filter) using gas

6 for each cooking style-(one Teflon and quartz fibre filter) using electricity

Pump used were calibrated with a gilibrator and the sampling flow was checked daily before and after every sampling exercise using a rotameter to ensure the flow rate.

Pumps used

- Cooking source- Diaphragm vacuum pump D7 DE Parallel 30l/min (attached to an adjustable flow meter of 30L/min)
- Personal monitoring- 182170B: APEX Lite (Standard) pump
- Microenvironment- Universal PCXR4 personal or area air sampling pump features a 5 to 5000 ml/min

Rotameter-

- Skc field rotameter cat no-320-4A5(0.4-5 L/min)
- SKC Field rotameter cat no-320-530(3-30L/min)
- Gilian Gilibrator-2 NIOSH Primary Standard Air Flow Calibrator

Cylone

- Personal monitoring at 3l/min to collect $PM_{2.5}$ with URG Teflon coated aluminium cyclone.
- For microenvironment monitoring a flow of 16.7l/min was use to collect $PM_{2.5}$.

2.1.2 Real kitchen sampling

Sampling was carried out in the kitchen of a residential house in Birmingham, with no other activities occurring during the cooking exercise in order to minimize the influence of emission from other indoor PM sources. The sampling took place between July and August 2014 and in October 2014. The kitchen was about 9meters by 4meters and had a four-burner gas stove connected to the city gas supply system. The sampling instrument was placed on an elevated platform with its port facing the burner (located 0.5 m from the pan and 1.5 m above the ground). Sampling was carried out with all the windows and door closed. Apart from the investigator, no other person was in the house during the course of the sampling. The kitchen was ventilated between each cooking experiment by opening the window. The cooking duration was about 40-70 minutes, with samples collected via two PM2.5 partisol inlet that operated at 16L/min (1 with Teflon filter- for gravimetric analysis and 1with a quartz fibre filter-for organic analysis)

After the sampling campaign the filters were stored in air tight metal tins and placed in the freezer of -22^{o}C.

Summary of samples collected-

- **Real kitchen samples- August campaign**-

  - Ambient samples during cooking 6 for each cooking style-(one Teflon and quartz fibre filter)

9 set of samples from grilling and cooking with ventilation (one Teflon and quartz fibre filter)

  - Micro environment 8 hrs and 16hours- quartz fibre filters for 10 days
  - 24 hours Personal monitoring during cooking (one Teflon and one quartz fibre filter)
  - Personal monitoring during no cooking – (total suspended particles and $PM_{2.5}$ and quartz fibre filters)

- **Real kitchen samples- October sampling**

➢ Micro environment- Two (2) $PM_{2.5}$ partisol inlet to operate at 16.7 L/min (SAMPLE FOR 8HOURS DURING COOKING)-

1 with Teflon filter- for gravimetric analysis
1-quartz fibre filter-for organic composition

➢ Personal monitoring-

3LPM-PM2.5(TEFLON) with cyclone
3LPM PM2.5 Quartz fibre filter for organic analysis

➢ Cooking source sample placed near the cooker-during each dish(about 60mins) with two (2) $PM_{2.5}$ partisol inlet to operate at 16L/min-

1 with Teflon filter- for gravimetric analysis
1-quartz fibre filter-for organic analysis

Figure 8 Sampling in home kitchen with extractor fan off ,A. without size selective inlet, B. with $PM_{2.5}$ inlet

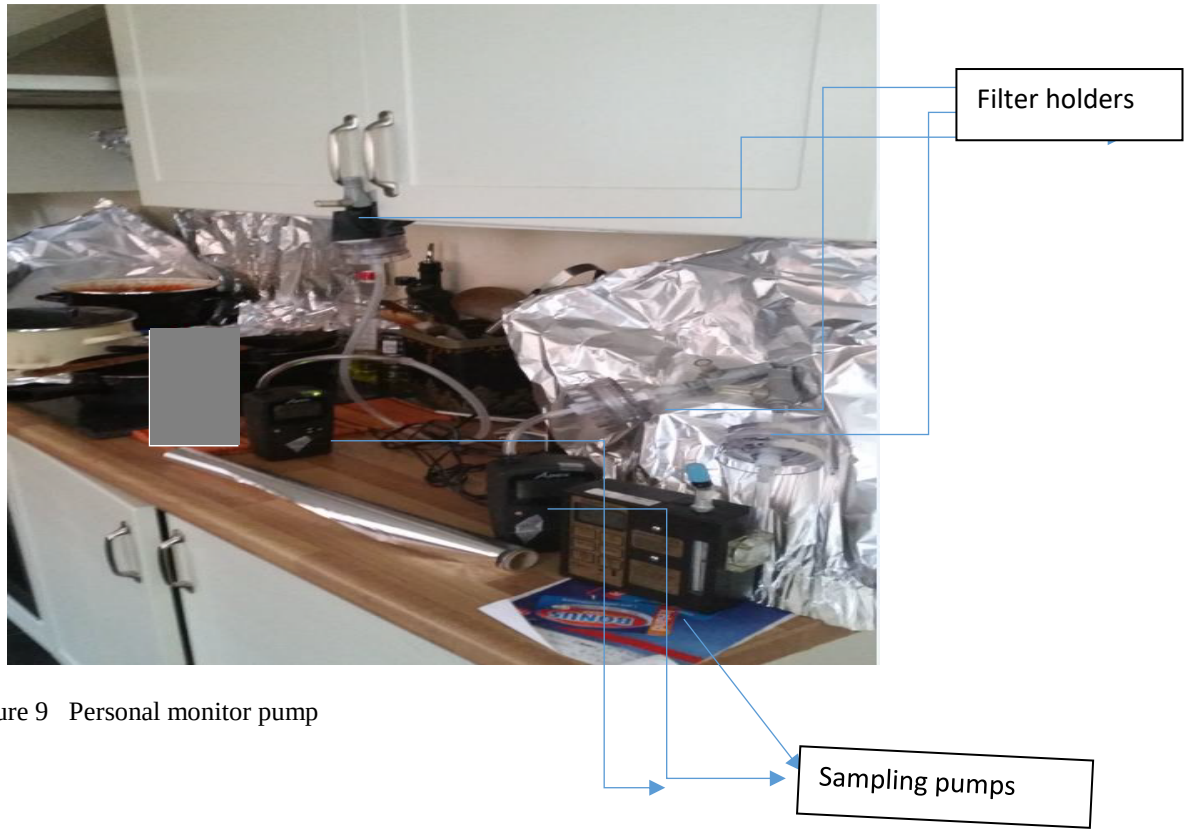


Figure 9 Personal monitor pump

In the kitchen the following instruments were used for sample collection

- Personal sampler pumps for 24hr personal exposure (1 teflon and 1 quartz fiber filter)
- Air sampling pumps for taking samples while cooking (1 teflon and 1 quartz fiber filter)
- Microenvironment sampling pump for taking 24 hour samples of the kitchen in two parts-a. during cooking activities and b. after cooking.

The food options cooked during the sampling was the same as that cooked in the trailer kitchen listed in Table 12.

Figure 10 Sampling in kitchen while grilling chicken.

COOK OFF PROJECT- A sampling campaign prroject between the University of Birmingham and University of Manchester (Cook off project) was carried out between 20th March 2012 and 22nd March 2012 where foods were cooked in the designed trailer laboratory kitchen described above**.** Different types of oils were also fried in glass beads to simulate the cooking process and emissions only associated to oils were analysed. During these cooking experiments, the Aerosol mass Spectrophotometer was collecting samples from the duct of the extractor pipe and anal

2.1.2.1 The Scanning Mobility Particle Sizers

The instrument, as shown in Figure 11, comprises of a TSI 3080 electrostatic classifier, a TSI 3081 differential mobility analyzer (long DMA) linked to a TSI 3022A condensation particle counter (CPC).

The particles entering the system are neutralized (using a radioactive source such as ^{85}Kr) such that they have a Fuchs equilibrium charge distribution and then, enter the Differential Mobility Analyser (DMA) where the aerosol is classified according to electrical mobility, with only particles of a narrow range of mobility exiting through the output slit. This monodisperse

distribution then goes to the Condensation Particle Counter which determines the particle concentration at that size.

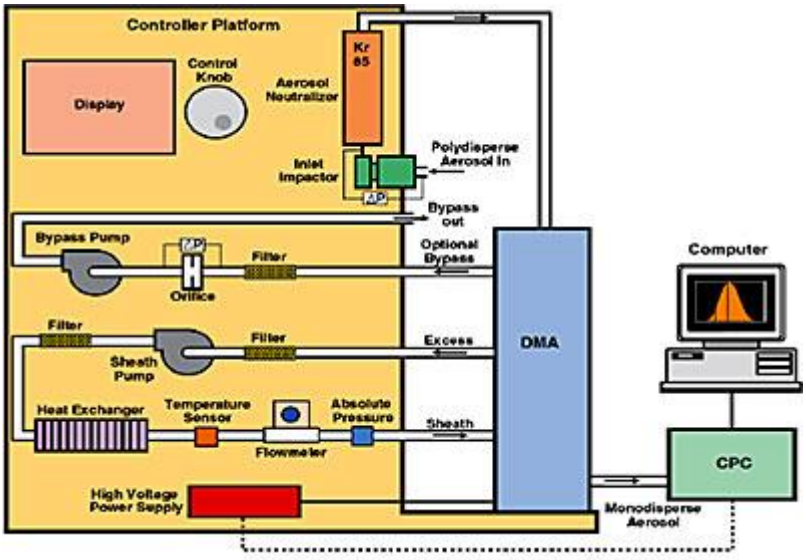


Figure 11 Schematic of SMPS (TSI, 2010)

2.1.2.2 The Differential Mobility Analyser (DMA) (TSI 3081)

The Differential Mobility Analyser (DMA) ,Figure 13 , contains an outer, grounded cylinder and an inner cylindrical electrode that is connected to a negative power supply. The electric field between the two concentric cylinders separates the particles according to their electrical mobility which is inversely related to the particle size. Particles with negative charge are repelled towards and deposited on the outer wall. Particles with neutral charge exit with the excess air. Particles with positive charge move rapidly towards the negatively-charged center electrode. Only particles within a narrow range of electrical mobility have the correct trajectory to pass through an open slit near the DMA exit. The electrical mobility of these selected particles is a function of flow rates, geometric parameters and the voltage of the center electrode.

2.1.2.3 The Condensation particle counter (TSI 3022A)

The Condensation particle counter, Figure 12, determines the total particle number concentrations by the growing of detected particles to larger sizes achieved by the condensation of supersaturated vapour (butanol). These particles are then counted using an optical laser detector. The TSI model 3025A can count particles of sizes >3 nm, and the model 3022A particles >7 nm and maximum detectable concentration is 10^5 cm^{-3} and10^7 cm^{-3} respectively. The CPC TSI 3025 operates with continuous aerosol flow which is saturated with butanol in a slightly heated saturator chamber and then the aerosol passes to the condenser where the temperature of the butanol-aerosol mixture is decreased by 17-27°C. The butanol become supersaturated and condense onto the particles, which grow to droplets of several µm in diameter, and are focused in a nozzle and introduced into a counting optic which is able to count every single particle.

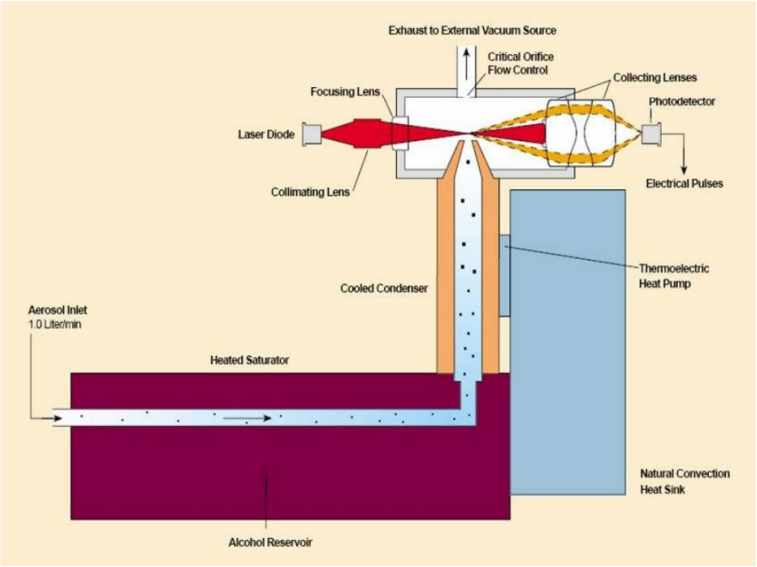


Figure 12 Schematic of CPC by TSI

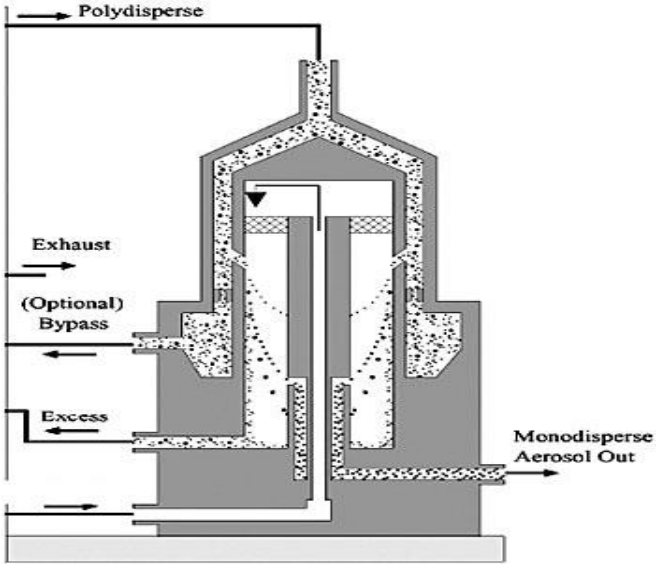


Figure 13 Schematic of DMA

2.1.3 Stratford Road Birmingham

The site is an urban roadside location located at the junction of Highgate Road (Balti Triangle) Figure 15. The area consists of numerous restaurants as shown in the map mainly of Indian cuisine.

Figure 14 Samplers kept in cabin owned by Birmingham city council with inlets placed on the roof of the cabin.

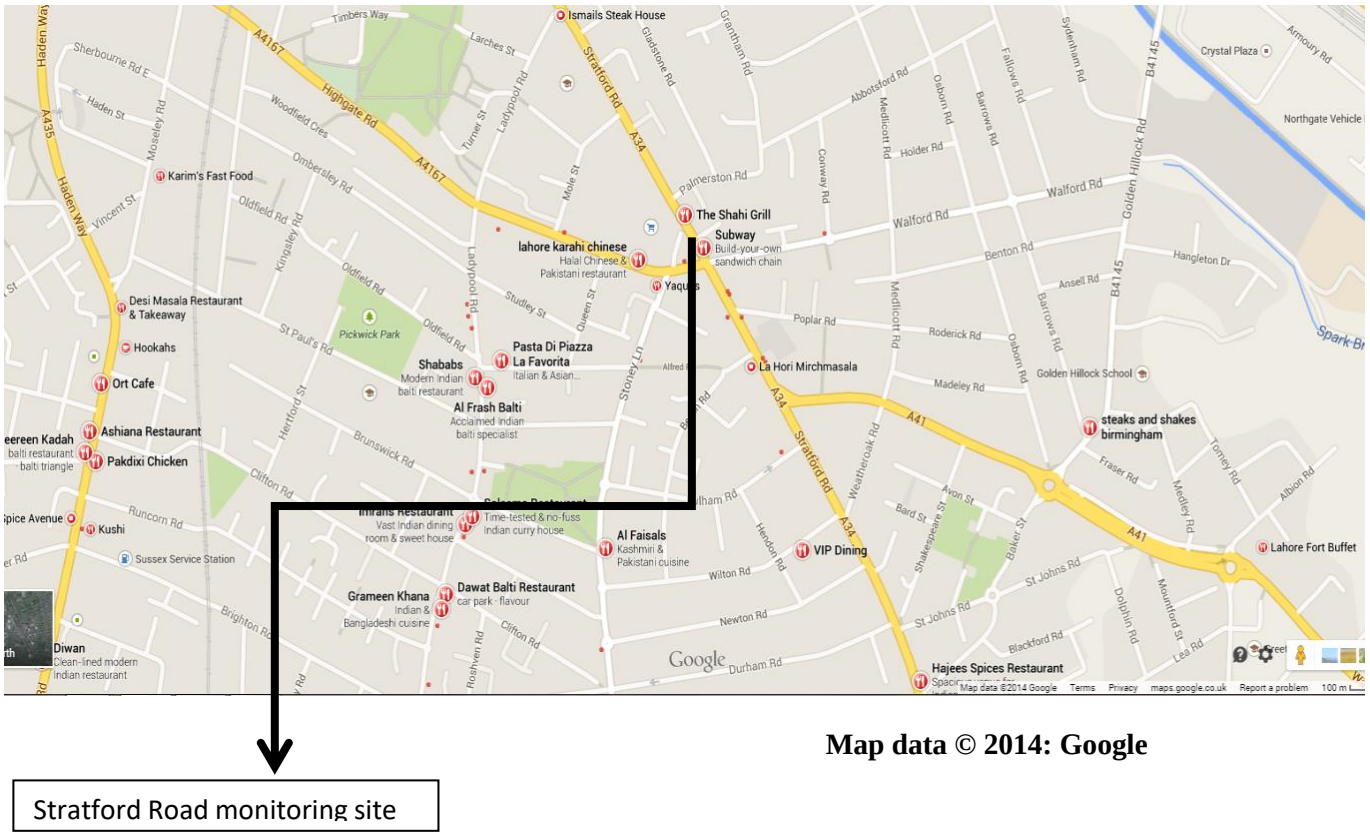


Figure 15 Stratford road map showing location of samplers in cabin owned by Birmingham city council and neighbouring restaurants. (Map data 2014: Google)

Figure 16 Hi-volume sampler and partisol sampler

2.1.3.1 Digitel high volume aerosol sampler

Digitel high-volume sampler (model DHA-80 Digitel Elektronik),Figure 17 and Figure 18, automatic air sampler for the collection of aerosol samples (PM2.5). The instrument uses the filtration/impaction method for collection of samples. During the sampling period the digitel operated at about 500 L min^{-1} collecting $PM_{2.5}$ on 150 mm quartz fiber filters held in holders. The instrument automatically switches and loads new filters after 24hours of sampling from filters placed in its inbuilt filter cassette stock (autosampling).

Analysis on filters collected- Organic and elemental carbon, Organic compound analysis.

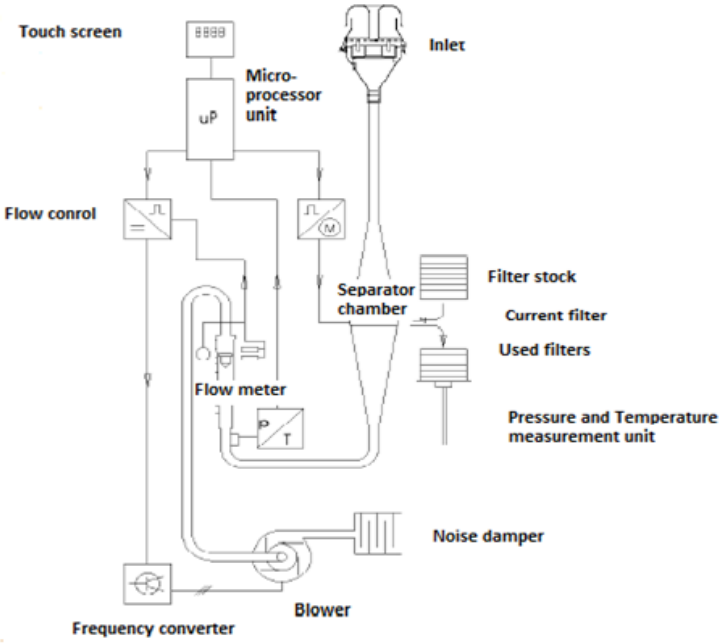


Figure 17 Hi-volume schematic unit of a Digitel high-volume sampler (Enviro Technology Services Plc, n.d)

Figure 18 Hi-volume sampler unit of a Digitel high-volume sampler (Enviro Technology Services Plc, n.d)

2.1.3.2 Partisol dichotomous sequential air sampler

A Partisol-Plus dichotomous sequential sampler (Model 2025), **Figure 19** and **Figure 20**, was used for 24-hour sampling of $PM_{2.5}$ and PM $_{2.5\text{-}10}$ on PTFE filters of diameter 47 mm. The Partisol air sampler splits a PM_{10} sample stream into $PM_{2.5}$ and PM $_{2.5\text{-}10}$ fractions with a

virtual impactor (Rupprecht & Patashnick Inc., 2001). The instrument consists of four cartridges each with a capacity of 16 cassettes containing the 47 mm filters. The two cartridges automatically supply the loaded 47mm filters for sampling while the remaining two cartridges store the exposed filters. The volumetric flowrate of the Partisol is 16.7 L min-1 for fine PM, and 1.7 L min-1 for coarse PM.

Figure 19 Partisol 2025 sampler (Rupprecht & Patashnick Inc., 2001)

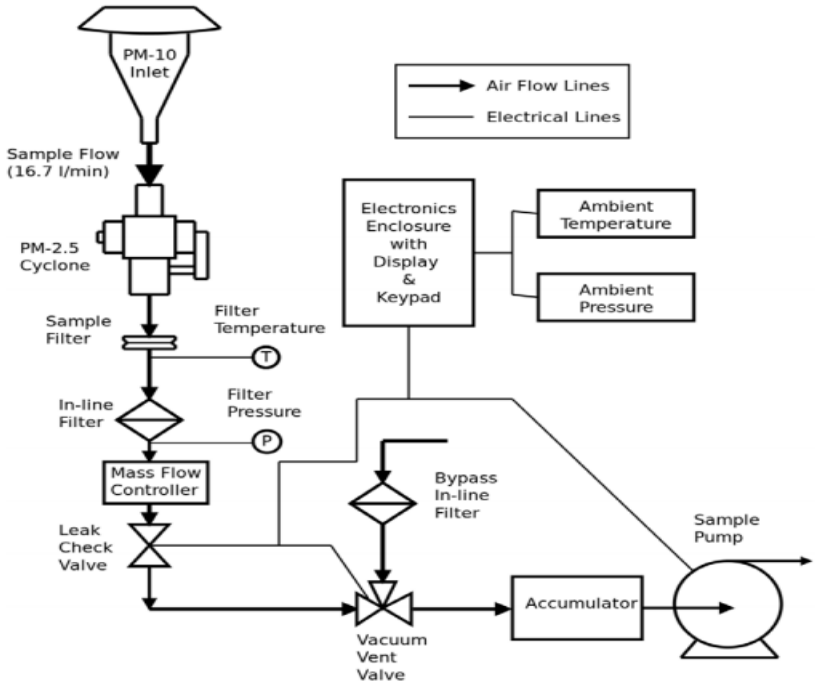


Figure 20 Partisol 2025 flow schematics (Rupprecht & Patashnick Inc., 2001)

The samples collected on filters undergo gravimetric determination and subsequent chemical analysis. Inert filters(filters made with chemically inert materials) such as Teflon and quartz fiber filters are used due to their nature as they will not present any interference to components collected on them. Teflon filters have low background levels of many analytes and are thus useful in low volume sampling applications.

Filter based method of measurement has the potential for positive and negative artefacts as possibility exists for the reaction of trace gases with particles on the filter or the filter itself resulting on positive artefact and also there could be some evaporative loss of semi-volatile components resulting in negative artefacts. The set out procedures and guidelines have to be followed strictly to keep these artefacts to the barest minimum.

After the particles are collected on the filters they are extracted with organic solvents followed by the quantification of organic compound concentration using gas chromatography/mass spectrometry (GC-MS).

2.2 Filter Preparation

Filters used

- QM-A quartz fibre Whatman 47mm
- PTFE membrane 47mm, Whatman

The quartz fibre filters were heated in a box furnace at filters were prebaked at 550^{o}C for 5 hours in to reduce blank carbon levels and then packed using aluminium foil and stored in a freezer at -20^{o} C awaiting sampling.

PTFE filters were weighed before and after sampling using a Sartorius Model MC5 microbalance (sensitivity- 1 μg). Before weighing, all filters were equilibrated in humidity (35-45% relative humidity) and temperature (20±1^{o}C) controlled windowless room for 24 hours before weighing commenced (Yin and Harrison, 2004). An ionizing blower and an α-particle source (210_{Po}) were used to reduce the effects of static electricity on the filter. Each filter was weighed three times and both positive and negative weights were recorded. Average weights were calculated using the arithmetic mean of the six recorded values.

Before the sampling episodes the filter holders are cleaned with distilled and deionized water and wrapped in clean foil immediately. After sampling exposed filters are placed in aluminium tins and wrapped in foil paper sealed in polyethene bags and stored in the freezer. The filters were weighed in the weighing room (after sampling) without 24hr exposure to prevent loss of volatile species collected from the sampling.

The quartz fibre filter samples were analysed for organic species, prior to extraction the filters were spiked with isotopically labeled internal standards for quantification, including octacosane-d58, hexatriacontane-d74, dibenz(ah)anthracene-d14, aaa-20R-cholestane-d4, heptadecanoic acid-d33, levoglucosan-U13C6 and cholesterol-2,2,3,4,4,6-d6. After extraction the samples were analysed using a gas chromatography mass spectrometry system (GC–MS) from Agilent Technologies (GC – 6890N plus MSD – 5973N) fitted with a HP-5MS column

(30 m, 0.25 mm diameter, 0.25 μm thickness). The extraction method was based upon(Sheesley et al., 2004) , acid derivatisation upon (Podlech, 1998) and (Aldai et al., 2005) and levoglucosan and cholesterol derivatisation upon(Yue and Fraser, 2004) (Yin et al., 2010)

Gravimetric analysis:-To determine the mass of particulate matter collected on teflon during sampling, the difference between the mass of the filter before and after sampling mass was calculated. PTFE filters were used for gravimetric analysis since they are not prone to absorption of ambient water vapour (Dikken, 2013).

Blank filters-

Blanks filters (which are filters that were prepared with the sampling filters and placed in a filter holder but not exposed or sampled upon) were taken along to the sampling location and collected during the sampling exercises with one blank filter collected for every 6 samples taken. Laboratory blanks (unused filter not taken to the field) were also collected. The blank field and laboratory filters were analysed with the sample batches, which included gravimetric analysis as well as other analysis such as organic extraction, OC/EC runs and XRF analysis and the samples were blank corrected.

2.3 Organic analysis.

GC–MS can analyse of polar compounds, such as sterols, dicarboxylic acids and fatty acids, involves derivatization procedure which results in the conversion of the organic acids to their esters, to reduce their polarity and increase the instrument response. This is generally labour intensive and time consuming as the methods need sample preparation such as extraction, concentration and pre-separation(Li et al., 2009).

2.3.1 Clean up procedures

- Glassware-The glassware used were washed and rinsed with deionized distilled water then heated in the oven at 520^{o}C for two hour. After they were allowed to cool down they were rinsed three times with dichloromethane before use.

- Each filter sample was then spiked with 50 µl of 10ppm internal standard mix-all (with composition listed as below).

Internal standards mix-all (ISALL)

1. octacosane-d58 (n-alkanes)
2. dibenz(ah)anthracene-14 (pahs)
3. heptadecanoic acid-d33 (fatty acids)
4. cholesterol-2,2,3,4,4,6-d6 (cholesterol)
5. Methyl-beta-D-xylopyranoside (levoglucosan)
6. aaa-20R-cholestane-d4 (hopanes)

2.3.2 Mild sonication

The filters were extracted at room temperature (25^{o}C) with 30 ml aliquots of DCM (dichloromethane, HPLC grade) twice and 30 ml aliquots of methanol (twice) under mild ultrasonic agitation for 15 minutes for each aliquot.

The extracts were combined and transferred to a turbo evaporator tube for concentration after the sonication. Three washes were done on the flask containing the filters after the second aliquot extract of each solvent, each using 2 pipettes full, with DCM and methanol respectively.

2.3.3 Concentration

The combined extract was reduced in volume to approximately 5 ml using the turbo evaporator equipment (at temperature 30^{o}C and pressure 0.5 bar for about 50 mins for DCM evaporation, and at 45^{o}C and 1.0 bar for about 100 mins for methanol evaporation).

The extract left was then transferred to a 10 ml clean calibrated graduated finger glass vial through a clean glass pipette column packed with glass wool and anhydrous Na_2SO_4 (1.0g) (the packed column was pre-rinsed with 2 pipettes of DCM before use). The turbo tube was washed four times with 2 pipettes DCM and 2 pipettes of methanol after the transfer into the 10ml glass vial. The packed column was then rinsed 2 times with DCM and methanol.

The sample was then concentrated under a nitrogen flow and during the blow down process additional DCM was added to prevent DCM depletion. The final extraction volume was 500 µl and 100 µl each of the concentrated solution was then poured into 5 different GCMS vials, covered tightly, sealed with paraffin tape and stored in the freezer at -18^{o}C.

2.3.4 Derivitization of extracts.

2.3.4.1 PROCEDURE FOR ORGANIC ACID METHYLATION.

2.0M trimethylsilyldiazomethane (TMS-DM) in diethyl ether was used to derivatise acids. 100 µl (10 pipette drops) of standard solution was placed into a 2 ml clean glass vial and 100 µl (10 pipette drops) of methanol was added followed by addition of 150 µl (15 pipette drops) of 2.0M TMS-DM reagent. With the vial open, it was shaken well and placed in a preheated dry heater bath at 40^{o}C for 10 minutes and then left at room temperature for another 50 minutes. The solution was then blown down to near dryness using nitrogen blower (making sure the product solution was not dried completely). 100 µl (10 pipette drops) of n-hexane was then added and

shaken well and transferred to a 100 µl GC vial followed by three washes (3 pipettes) then concentrated down to 100 µl (original volume) before being subjected to GC analysis.

2.3.4.2 PROCEDURE FOR DERIVATISATION OF STEROLS.

Samples derivatised using the procedure below to convert the sterols to their trimethyl silyl ether.

N,O-Bis(trimethylsilyl)trifluoroacetamide plus 1% trimethylchlorosilane (BSTFA-TMCS) was used to derivatise cholesterol, levoglucosan, stigmesterol and their standards. The procedure followed was 100 µl (10 pipette drops) of standard solution was transferred to a GC-MS vial and concentrated down to near dryness (ensuring that the solution was not dried completely). 100 µl (10 pipette drops) of BSTFA-TMCS (99%:1%) was then added to the vials containing the samples. The vials were then capped and heated on a dry heater block at 80^{o}C for 1 hour. Then the vials were allowed to cool in a desiccator for 1 hour and subsequently subjected immediately to GC-MS analysis.

2.4 GCMS analysis

The organic compounds contained in extracts were analysed using a Gas Chromatography Mass Spectrometer (GCMS).

Principle of the GCMS- Based on the difference in the chemical properties of different molecules contained extracts, molecules separate as the sample travels the length of the GCMS column. Different retention time of molecules lead to them being eluted at different times from the gas chromatograph allowing the mass spectrometer downstream to capture, ionize, accelerate, deflect, and detect the ionized molecules separately. The mass spectrometer breaks each molecule into ionized fragments and fragments are detected by their mass to charge ratio. The compounds are identified in

the sample when identifying mass spectrum appear at characteristic retention times in the GC-MS analysis of the sample.

Prior to extraction of the quartz fibre filter samples, the filters were spiked with isotopically labeled internal standards for quantification, including octacosane-d58, hexatriacontane-d74, , dibenz(ah)anthracene-d14, aaa-20R-cholestane-d4, heptadecanoic acid-d33, , levoglucosan-U13C6 and cholesterol-2,2,3,4,4,6-d6. After extraction the samples were analysed using a gas chromatography mass spectrometry system (GC–MS) from Agilent Technologies (GC – 6890N plus MSD – 5973N) fitted with a HP-5MS column (30 m, 0.25 mm diameter, 0.25 μm thickness). The extraction method was based upon(Sheesley et al., 2004) , acid derivatisation upon (Podlech, 1998) and (Aldai et al., 2005) and levoglucosan and cholesterol derivatisation upon(Yue and Fraser, 2004,Yin et al., 2010).

Preparation of the standards purchased which were used for calibration of the GCMS as well as internal standards for the cooking sample collected. Below are a list of tables of the compounds which were analysed for with the GCMS (Table 13), standards purchased, and the initial stock concentrations prepared.

Table 13 Compounds for analysis(Linstrom and Mallard, 2012)

Compound	**molecular mass**	**Target ion**	**qualifier 1**	**qualifier 2**	**qualifier 3**	**formula**
N-Alkanes	**g/mol**	**m/z**	**m/z**	**m/z**	**m/z**	
n-tetradecane	198	57	43	71	85	$C_{14}H_{30}$
n-pentadecane	212	57	43	71	85	$C_{14}H_{30}$
n-hexadecane	226	57	43	71	85	$C_{16}H_{34}$
n-heptadecane	241	57	43	71	85	$C_{17}H_{36}$
n-octadecane	255	57	43	71	85	$C_{18}H_{38}$
n-nonadecane	269	57	43	71	85	$C_{19}H_{40}$
n-eicosane	283	57	43	71	85	$C_{20}H_{42}$
n-heneicosane	297	57	43	71	85	$C_{21}H_{44}$
n-docosane	311	57	43	71	85	$C_{22}H_{46}$
n-tricosane	325	57	43	71	85	$C_{23}H_{48}$
n-tetracosane	339	57	71	43	85	$C_{24}H_{50}$
n-pentacosane	353	57	71	43	85	$C_{25}H_{52}$

n-hexacosane	367	57	71	43	85	$C_{26}H_{54}$
n-heptacosane	381	57	43	71	85	$C_{27}H_{56}$
n-octacosane	395	57	71	43	85	$C_{28}H_{58}$
n-nonacosane	409	57	71	43	85	$C_{29}H_{60}$
n-triacontane	423	57	71	85	43	$C_{30}H_{62}$
n-hentriacontane	437	57	43	71	85	$C_{31}H_{64}$
n-tritriacontane	465	57	71	43	85	$C_{33}H_{68}$
Polycyclic Aromatic Hydrocarbons						
benzo[ghi]fluoranthene	226	226	113	224		$C_{18}H_{10}$
chrysene	229	228	226	229		$C_{18}H_{12}$
benzo[b]fluoranthene	252	252	253	250		$C_{20}H_{12}$
benzo[k]fluoranthene	252	252	253	250		$C_{20}H_{12}$
benzo[a]fluoranthene	252	252				$C_{20}H_{12}$
benzo[b]pyrene	252	252				$C_{20}H_{12}$
benzo[a]pyrene	252	252	253	250		$C_{20}H_{12}$
perylene	252	252	253	250	126	$C_{20}H_{12}$
indeno[1,2,3-c,d]pyrene	276	276	277	274		$C_{22}H_{12}$
benzo[ghi]pyrene	276	276				
coronene	300	300	301	298		$C_{24}H_{12}$
acephenanthrylene	202	202				$C_{16}H_{10}$
benzo[e]pyrene	252	252	250	253		$C_{20}H_{12}$
Benzo[a]anthracene	228	228	226	229		$C_{18}H_{12}$
N-Alkanoic acids		**(methyl ester)**				
hexanoic acid	116	60	73	41		$C_6H_{12}O_2$
heptanoic acid	130	60	73	41	43	$C_7H_{14}O_2$
octanoic acid	144	60	73	43		$C_8H_{16}O_2$
nonanoic acid	158	60	73	57	41	$C_9H_{18}O_2$
decanoic acid	172	60	73	41	43	$C_{10}H_{20}O_2$
undecanoic acid	186	60	73	43	41	$C_{11}H_{22}O_2$
dodecanoic acid	200	73	60	43		$C_{12}H_{24}O_2$
tridecanoic acid	214	73	60	43		$C_{13}H_{26}O_2$
tetradecanoic acid (myristic acid)	228	73	60	43		$C_{14}H_{28}O_2$
pentadecanoic acid	242	73	43	60		$C_{15}H_{30}O_2$
hexadecanoic acid (palmitic acid)	256	43	73	60		$C_{16}H_{32}O_2$
heptadecanoic acid	271	73	60	57	43	$C_{17}H_{34}O_2$
octadecanoic acid (stearic acid)	285	43	73	60	57	$C_{18}H_{36}O_2$
nonadecanoic acid	299	43	73	57	60	$C_{19}H_{38}O_2$
eicosanoic acid	313	43	57	73		$C_{20}H_{40}O_2$
docosanoic acid	341	340	57	73		$C_{22}H_{44}O_2$
tetracosanoic acid	369	43	57	73		$C_{24}H_{48}O_2$
Unsaturated Fatty Acids		**(methyl ester)**				
9-hexadecenoic acid (palmitoleic acid)	254	55	41	69		$C_{16}H_{30}O_2$
9,12-octadecadienoic acid (Linoleic acid)	280	67	81	55	41	$C_{18}H_{32}O_2$

9-octadecenoic acid (oleic acid)	282	69	83			$C_{18}H_{34}O_2$
Dicarboxylic Acids		**(methyl ester)**				
butanedioic acid	118	55	45	74		$C_4H_6O_4$
pentanedioic acid	132	86	42	44		$C_5H_8O_4$
hexanedioic acid	146	100	43	60	41	$C_6H_{10}O_4$
heptanedioic acid	160	55	60	83		$C_7H_{12}O_4$
octanedioic acid	174	138	69	97	60	$C_8H_{14}O_4$
nonanedioic acid	188	55	41	60		$C_9H_{16}O_4$
decanedioic acid	202	98	55	60	41	$C_{10}H_{18}O_4$
undecanedioic acid	216	98	84	55		$C_{11}H_{20}O_4$
Monosaccharide Anhydrides		**(trimethyl silyl ether)**				
levoglucosan	162	204	217	333		$C_6H_{10}O_5$
Sterols						
cholesterol	387	43	55	57		$C_{27}H_{46}O$
Other Compounds						
benzoic acid	122	105	122	77		$C_7H_6O_2$

2.4.1 Standard preparations.

The standard solutions were prepared using the stock standard solutions Listed in table A, below, (of 2-0.1mg/ml for the various deuterated standards.

As a first step, 10ppm (10,000pg/µl) of each standard was prepared in 1000ul solution using the following formula:

$C_1V_1=C_2V_2$……..(1)

WHERE

C represents the concentration of solution 1(before) and 2(after) dilution.

V represents volume of standard 1(before) and 2(after) dilution

Using formula 1 table 2

Table A. Internal standard and recovery standard stock used.

ID	Compound	Abreviation	solvent	STOCK CONCENTRATION	
				mg/mL	pg/µL
ISA	octacosane-d58	Oct d58	Isooctane	1	1000000
ISD	dibenz(ah)anthracene-d14	D(ah)A d14	Toluene	0.1	100000
ISG	heptadecanoic acid-d33	HepDea d33	Isooctane	1	1000000
ISH	Methyl-beta-D-xylopyranoside	MXP	Methanol	1	1000000
ISI	cholesterol-2,2,3,4,4,6-d6	Chol d6	Methanol	1	1000000
RECOVERY STANDARD	p-terphenyl-d14	PTPd14	Isooctane	2	2000000

10ppm IS1, IS2, IS3, IS4 & IS5 (1ml) preparation					
IS Level	ID	Compound	VOL. IS Stock	VOL.	Final Concentration
New Standard Name	STOCK		Stock Solution V	SOLVENT Volume (ul)	pg/uL
IS1	ISA	octacosane-d58	10	Isooctane (990)	10,000
IS2	ISD	dibenz(ah)anthracene-d14	100	Toluene (900)	10,000
IS3	ISG	heptadecanoic acid-d33	10	Methanol (990)	10,000
IS4	ISH	Methyl-beta-D-xylopyranoside	10	Methanol (980)	10,000
	ISI	cholesterol-2,2,3,4,4,6-d6	10		10,000
IS5	ISB	2-Nonanone-1,1,1,3,3-d5 98 atom % D	10	Methanol (990)	10,000
RECOVERY STANDARD	SOLN X	p-terphenyl-d14	5	isooctane (995)	10,000

- Using formula 1, the spiking standard consisting of the deuterated compounds with concentration of 5000 and 10000pg/µl was prepared. The volume, V_2 of each stock standards A,D,G,H,I,J to be measured and made up to 1000µl was calculated. The final standard prepared shall be used for spiking of filters before extraction.

Spiking standard - To be spiked on the filter.

Stock solution	Conc 10/5ppm	VoL of IS STOCK Solution (ul)	DCM+METHANO	TOTAL VOL ISALL (ul)	Final concentration	
	compounds	v2			pg/ul	
ISA	octacosane-d58	5	905	1000	5,000	
ISD	dibenz(ah)anthracene-d14	50			5,000	
ISG	heptadecanoic acid-d33	10			10,000	
ISH	Methyl-beta-D-xylopyranoside	10			10,000	
ISI	cholesterol-2,2,3,4,4,6-d6	10			10,000	
ISJ	2-Nonanone-1,1,1,3,3-d5 98 atom % D	10			10,000	

The working standard for calibration of the GCMS were prepared so that the concentration of compounds will be from 5000pg/µl-0pg/µl (5000, 2000, 1000, 500, 200,50,20 and 0) recovery standard concentration of 1000 pg/µl and internal standard concentration of 500pg/µl for alkanes , hopanes and PAHs and 1000pg/µl for acids and sterols.

2.4.2 GCMS calibration

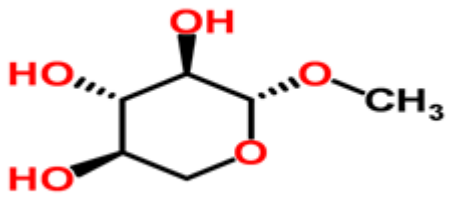
Natural standards were prepared with a range of concentrations ranging from 0 to 5000pg/µl, and calibration curves were prepared for the range of compounds.

CALIBRATION CURVE.

A calibration curve is a plot of the response ratio (STD natural response against the corresponding deuterated STD response) against the amount ratio (ratio of concentration of standard eg- 0-10,000 pg/µL to concentration of internal standard 1000pg/µl).

Methyl β-D-xylopyranoside was used as the internal standard for Levoglucosan and it is observed from the structure that there is no deuterium in the compound. The reason for selection of this compound as the internal standard for levoglucosan is as a result of their similar breakdown products from silyation. Levoglucosan (1,6-anhydro-β-D-glucopyranose, CAS number 498-07-7) is typically characterized by its base peak at *m*/*z* 204 and by *m*/*z* 217 and/or

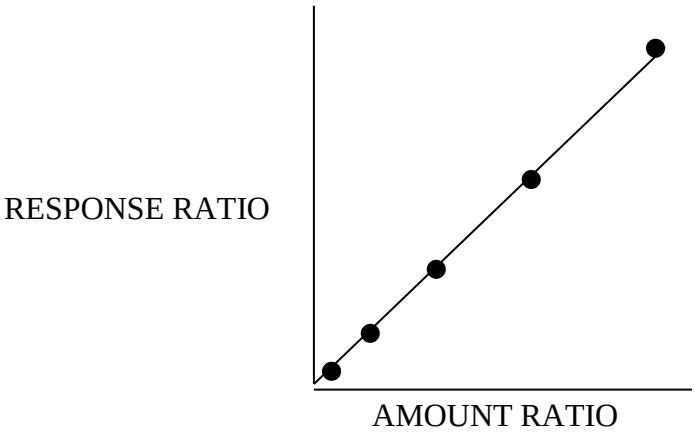
m/*z* 333 in mass fragmentograms of derivatized (silylated) samples while derivatized (silylated) Methyl β-D-xylopyranoside (CAS Number 612-05-5) are by its base peak at m/z 217, 204. The mass spectrum of levoglucosan trimethylsilyl ether exhibits only a small molecular ion (*m*/*z* 378) with fragments due to loss of CH_5Si (*m*/*z* 333), $C_6H_{17}OSi_2$ (*m*/*z* 217) and $C_7H_{18}OSi_2$ (*m*/*z* 204, base peak) while that of Methyl β-D-xylopyranoside is similar but at m/z 217 and 204 showing they break down similarly during derivatisation (Simoneit and Elias, 2001). The GC elution order is the factor that differentiates these compounds.



STRUCTURE FOR Methyl β-D-xylopyranoside-

As the only Methyl β-D-xylopyranoside present in the sample shall be that introduced during spiking, this standard can be used to quantify the amount of levoglucosan present in the samples analysed.

Calibration curve



As the calibration standard for the compound (0-G) are run in the GCMS, each produces a response for the compound investigated which is compared to its corresponding deuterated standard response producing a response ratio. This is then plot against its corresponding amount ratio to produce the calibration curve.

Linear regression is carried out on the plot to establish the equation that best describes the linear relationship between instrument response and analyte level. The relationship is described by the equation of the line, *i.e.*, $y = mx + c$, where m is the gradient of the line and c is its intercept with the y-axis. Where the instrument response is (y) and analyte level is (x) (LGC, 2003). The correlation coefficient, r (and the related parameters r^2) is a measure of the strength of the degree of correlation between the y and x values and the closer the r^2 is to 1, the stronger the correlation.

GCMS system used was Agilent GC- 6890N plus MSD-5973N fitted with a HP-5MS -30 m, 0.25 mm diameter, 0.25 µm thickness column.

CALIBRATION OF STEROLS.

Calibration standards 0-G are prepared from natural standard of sterols with concentrations ranging from 0 pg/µL to 25000 pg/µL with concentration of internal standard and recovery standards of 1000 pg/µL.

These varying concentrations are derivatised using the procedure below to convert the sterols to their trimethyl silyl ether, which is then analysed to produce a calibration curve.

<u>PROCEDURE FOR DERIVATISATION OF STEROLS.</u>

N,O-Bis(trimethylsilyl)trifluoroacetamide plus 1% trimethylchlorosilane (BSTFA-TMCS) was used to derivatise cholesterol, levoglucosan, stigmesterol and their standards. The

procedure followed was 100 µl (10 pipette drops) of standard solution was transferred to a GC-MS vial and concentrated down to near dryness (ensuring that the solution was not dried completely). 100 µl (10 pipette drops) of BSTFA-TMCS (99%:1%) was then added to the vials containing the samples. The vials were then capped and heated on a dry heater block at 80^{o}C for 1 hour. Then the vials are allowed to cool in a desiccator for 1 hour and subsequently subjected immediately to GC-MS analysis.

ACIDS CALIBRATION CURVES

Natural standards for acids were then prepared with a range of concentrations ranging from 0 to 10000pg/µl, with known concentration of internal standard (1000pg/µl) and recovery standards (1000pg/µl). These levels of concentrated solution were derivatised and then run on the GCMS and the calibration graphs were plotted.

PROCEDURE FOR ORGANIC ACID METHYLATION.

2.0M trimethylsilyldiazomethane (TMS-DM) in diethyl ether was used to derivatise acids. 100 µl (10 pipette drops) of standard solution was placed into a 2 ml clean glass vial and 100 µl (10 pipette drops) of methanol was added followed by addition of 150 µl (15 pipette drops) of 2.0M TMS-DM reagent. With the vial open, it was shaken well and placed in a preheated dry heater bath at 40^{o}C for 10 minutes and then left at room temperature for another 50 minutes. The solution was then blown down to near dryness using nitrogen blower (making sure the product solution was not dried completely). 100 µl (10 pipette drops) of n-hexane was then added and shaken well and transferred to a 100 µl GC vial followed by three washes (3 pipettes) then concentrated down to 100 µl (original volume) before being subjected to GC analysis.

MONOGYCERIDE CALIBRATION CURVES.

Similar to other sets of compounds, calibration standards 0-G are prepared from natural standard of monoglycerides with concentrations ranging from 0=0 pg/µL to G=25000 pg/µL with concentration of internal standard and recovery standards of 1000 pg/µL.

These varying concentrations were derivatised using the procedure used above to convert sterols to their trimethyl silyl ether. The derivatised standards were then analysed to produce calibration curves.

DEFINITIONS

Detection limits.

- The instrument detection limit (IDL) is defined as the amount of pollutant that gives a signal to noise ratio of 3 : 1, was determined by calculating the signal to noise ratio for the pollutant in the lowest calibration standard (Harrad, 2005). It is the lowest analyte concentration for feasible instrument detection.

$$IDL = Conc \times \frac{3}{SNR}$$

Where *IDL*=instrument detection limit

Conc=concentration of target pollutant in calibration standard

SNR=signal to noise ratio obtained for that pollutant.

- The limit of quantification has been defined as the lowest concentration at which the analyte can not only be reliably detected but at which some predefined goals for bias and imprecision are met(Armbruster and Pry, 2008).

Limit of quantification = 10*standard deviation of blank ..…(1)

- The method detection limit is the minimum concentration of a substance that can be measured and reported with 99-percent confidence that the analyte concentration is greater than zero. It usually considers errors that could arise due to all the steps of the analysis.

Method Detection limit= 3*standard deviation of blank ..……(2)

The IDL for the GCMS instrument was carried out and for the compound was found to be as shown in Table 14.

Table 14 Instrument detection limit

Compound	IDL (pg/ul)
Alkanes	
Tetracosane	**4.1**
Pentacosane	**5.4**
Hexacosane	**4.5**
Heptacosane	**4.7**
Octacosane	**6.3**
Nonacosane	**7.3**
Triacontane	**9.0**
Hentriacontane	**10.5**
Dotriacontane	**8.5**
Tritriacontane	**10.7**
Tetratriacontane	**13.0**
Pentatriacontane	**13.3**
Polycyclic Aromatic Hydrocarbons	
Benzo[b]fluoranthene	**1.9**
Benzo[k]fluoranthene	**7.2**
Benzo[e]pyrene	**1.4**
Benzo[a]pyrene	**7.0**
Perylene	**2.4**
Indeno[123-cd]pyrene	**2.0**
Dibenzo[a,i]phenanthrene	**2.5**
Picene	**6.4**
Benzo[ghi]perylene	**1.8**
Coronene	**2.3**
Sterols	
levoglucosan	**0.9**
cholesterol	**2.4**
stigmasterol	**2.0**
Monoglycerides	

1-Monopalmitin	8.3
1-Monostearin	13.0
1-Monomyristin	9.1
1-Monoolein	16.7

Concentrations of compounds

Concentration of the various compounds was calculated using the following formula:

This was calculated using formula 5;

$$C = \frac{C_{NAT} \times V_{ex}}{V_{ins}} \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (3)$$

Where

Conc=concentration in sample

C_{NAT} =concentration of native compound in the sample (conc from GCMS)

V_{ex} =final volume of extract used for analysis

V_{ins} =Instrument sampling volume

SPIKED FILTER FOR PAH AND ALKANES

5 blank filter papers were prepared and extracted as described above, after being spiked with 50µl of 1000pg/µl natural standard solution. The extracts were subjected to GCMS analysis and the concentrations obtained are presented in Table 15.

- INTERNAL STANDARD RECOVERIES- was determined by using the ratio of internal standard peak areas and recovery determination standard peak areas in the samples and in the calibration standard. The average of values obtained from running calibration standard D (for both PAH and Alkanes).

$$\left(\left(\frac{A_{IS}}{A}_{RDS}\right)S * \left(\frac{A_{RDS}}{A_{IS}}\right)STD * \left(\frac{C_{IS}}{C}_{RDS}\right)S * \left(\frac{C_{RDS}}{C_{IS}}\right)STD\right) * 100 \ldots\ldots\ldots\ldots (4)$$

WHERE-

$\left(\frac{A_{IS}}{A_{RDS}}\right)S$ --IS RATIO OF INTERNAL STANDARD PEAK AREA TO RECOVERY DETERMINATION STD PEAK AREA IN SAMPLE.

$\left(\frac{A_{RDS}}{A_{IS}}\right)STD$--IS RATIO OF RECOVERY DETERMINATION STD PEAK AREA TO INTERNAL STANDARD PEAK AREA TO IN THE CALIBRATION STANDARD.

$\left(\frac{C_{IS}}{C_{RDS}}\right)S$-- IS RATIO OF CONCENTRATION OF RECOVERY DETERMINATION STD TO CONCENTRATION OF INTERNAL STANDARD IN SAMPLE.

$\left(\frac{C_{RDS}}{C_{IS}}\right)STD$ --IS RATIO OF CONCENTRATION OF INTERNAL STANDARD TO CONCENTRATION OF RECOVERY DETERMINATION STD IN CALIBRATION STANDARD.

For the natural standards prepared, as the final volume was 250µl and 50µl of the solutions were spiked on the filter, the concentration in the final extract is therefore 200pg/µl for the 1000pg/µl solutions.

Table 15 Spiked filter extract concentrations.

	SS1	SS2	SS3	SS4	SS5	AVERAGE	STD DEV	PRECISION	%REC
ALKANES	CONC pg/ul								
Tetracosane	306.53	311.44	265.38	294.14	309.48	297.4	19.1	6.4	149
Pentacosane	271.49	269.85	240.85	262.04	249.65	258.8	13.2	5.1	129
Hexacosane	308.66	278.64	280.84	264.43	273.13	281.1	16.6	5.9	141
Heptacosane	325.71	259.17	269.93	263.68	255.48	274.8	29.0	10.5	137
Octacosane	326.78	240.57	270.98	257.05	251.94	269.5	33.9	12.6	135
Nonacosane	341.64	254.12	264.42	243.82	250.5	270.9	40.2	14.9	135
Triacotane	325.38	227.62	242.79	209.98	213.42	243.8	47.4	19.4	122
Hentriacotane	333.16	206.59	217.43	207.72	198.8	232.7	56.5	24.3	116
Dotriacotane	329.47	136.9	184.92	159.96	155.97	193.4	77.9	40.3	97
Tritriacotane	231.76	112.47	142.39	126.83	142.17	151.1	46.8	30.9	76
Pentatriacotane	30.4	29.99	36.98	26.15	25.3	29.8	4.6	15.5	15
PAHs	CONC pg/ul								
Benzo[b]fluoranthene	151.32	131.01	126.87	131.91	144.26	135.3	10.3	7.6	68
Benzo[k]fluoranthene	235.69	207.45	215.74	224.08	261.01	220.7	20.8	9.4	110
Benzo[e]pyrene	165.67	157.3	159.28	166.47	183.08	162.2	10.2	6.3	81
Benzo[a]pyrene	162.71	155.97	159.95	155.65	183.86	158.6	11.7	7.4	79
Perylene	169.07	176.61	168.52	165.62	179.71	170.0	6.0	3.5	85
Indeno[123-cd]pyrene	162.36	156.67	172.84	159.93	175.38	163.0	8.2	5.0	81
Dibenz[ah]anthracene	223.04	202.08	193.83	232.17	177.29	212.8	22.1	10.4	106
Picene	236.5	230.46	234.22	262.38	243.15	240.9	12.6	5.2	120
Benzo[ghi]perylene	159.16	161.82	180.57	166.72	161.84	167.1	8.6	5.1	84
Coronene	165.73	177.73	183.05	199.87	188.85	181.6	12.7	7.0	91

WHERE PRECISION =((STANDARD DEVIATION/ MEAN)*100

To obtain the internal standard recoveries, natural standard D with a concentration of 500pg/µl for both PAHs and Alkanes was used. This standard was run 5 times to obtain average response for all the compounds, internal standard and recovery standard. This was used with equation 1, in section 2.2 to obtain the internal standard recoveries.

They ranged between 100 and 73%. 0 shows the average and standard deviation of the concentrations obtained, the standard deviation was found to be between 22 and 6 for PAHS and much larger for alkanes(13-77).

Table 16 PAH internal standard and natural standard recovery calculated.

	1000PG/UL							
SS1					Average Response of Standards (from vials)			
Target Compounds	**IS Name**	**IS RESP**	**Cmpd Resp**		**Average Resp IS**	**Average Resp NS**	%IS RECOVERY	% Nat Standard Recovery
Benzo[b]fluoranthene	p-Terphenyl-d14	388315	28038		183120.6	45121.8	89.5	73.3
Benzo[k]fluoranthene	Dibenz[ah]anthracene-d14	55905	45082		29470.4	74628		71.2
Benzo[e]pyrene			39597			81905		57.0
Benzo[a]pyrene			7047			26026.8		31.9
Perylene			11952			43289		32.6
Indeno[123-cd]pyrene			16253			34794.2		55.1
Dibenz[ah]anthracene			23278			34490.6		79.6
Picene			5142			8308.8		73.0
Benzo[ghi]perylene			17763			35674.8		58.7
Coronene			34846			59607.6		68.9
SS2					Average Response of Standards (from vials)			
Target Compounds	**IS Name**	**IS RESP**	**Cmpd Resp**		**Average Resp IS**	**Average Resp NS**		
Benzo[b]fluoranthene	p-Terphenyl-d14	340732	22577		183120.6	45121.8	101.3	67.2
Benzo[k]fluoranthene	Dibenz[ah]anthracene-d14	55568	35677		29470.4	74628		64.2
Benzo[e]pyrene			36195			81905		59.4
Benzo[a]pyrene			5340			26026.8		27.6
Perylene			14226			43289		44.2
Indeno[123-cd]pyrene			15067			34794.2		58.2
Dibenz[ah]anthracene			19112			34490.6		74.5
Picene			4779			8308.8		77.3
Benzo[ghi]perylene			18168			35674.8		68.4
Coronene			37135			59607.6		83.7
SS3					Average Response of Standards (from vials)			
Target Compounds	**IS Name**	**IS RESP**	**Cmpd Resp**		**Average Resp IS**	**Average Resp NS**		
Benzo[b]fluoranthene	p-Terphenyl-d14	254314	14676		183120.6	45121.8	92.7	58.6
Benzo[k]fluoranthene	Dibenz[ah]anthracene-d14	37933	26184		29470.4	74628		63.2
Benzo[e]pyrene			25218			81905		55.4
Benzo[a]pyrene			4316			26026.8		29.9
Perylene			7993			43289		33.2
Indeno[123-cd]pyrene			12397			34794.2		64.1
Dibenz[ah]anthracene			11965			34490.6		62.4
Picene			3404			8308.8		73.7
Benzo[ghi]perylene			14864			35674.8		75.0
Coronene			26106			59607.6		78.8
SS4					Average Response of Standards (from vials)			
Target Compounds	**IS Name**	**IS RESP**	**Cmpd Resp**		**Average Resp IS**	**Average Resp NS**		
Benzo[b]fluoranthene	p-Terphenyl-d14	288033	16509		183120.6	45121.8	86.8	58.2
Benzo[k]fluoranthene	Dibenz[ah]anthracene-d14	40215	29712		29470.4	74628		63.3
Benzo[e]pyrene			28703			81905		55.7
Benzo[a]pyrene			3807			26026.8		23.2
Perylene			7820			43289		28.7
Indeno[123-cd]pyrene			11356			34794.2		51.9
Dibenz[ah]anthracene			18014			34490.6		83.0
Picene			4730			8308.8		90.5
Benzo[ghi]perylene			13829			35674.8		61.6
Coronene			30211			59607.6		80.6
SS5					Average Response of Standards (from vials)			
Target Compounds	**IS Name**	**IS RESP**	**Cmpd Resp**		**Average Resp IS**	**Average Resp NS**		
Benzo[b]fluoranthene	p-Terphenyl-d14	206303	12859		183120.6	45121.8	82.7	63.2
Benzo[k]fluoranthene	Dibenz[ah]anthracene-d14	27450	26180		29470.4	74628		77.8
Benzo[e]pyrene			22692			81905		61.5
Benzo[a]pyrene			6039			26026.8		51.5
Perylene			7505			43289		38.5
Indeno[123-cd]pyrene			9211			34794.2		58.7
Dibenz[ah]anthracene			7089			34490.6		45.6
Picene			2706			8308.8		72.3
Benzo[ghi]perylene			8976			35674.8		55.8
Coronene			19487			59607.6		72.5

Table 17 Alkane internal standard and natural standard recovery calculated.

	1000PG/UL							
SS1					rage Response of Standards (from v			
Target Compounds	**IS Name**	**IS RESP**	**Cmpd Resp**		**Average Resp IS**	**Average Resp NS**	%IS RECOVERY	% Nat Standard Recovery
Tetracosane	p-Terphenyl-d14	126690	6650		198028.8	22666.4	**87.0**	34.6
Pentacosane	Octacosane-d58	8052	4801		14460.8	20406.6		27.7
Hexacosane			5450			17674.6		36.4
Heptacosane			6268			18257.2		40.5
Octacosane			5916			16447.8		42.4
Nonacosane			5963			14558.6		48.3
Triacotane			4948			12149		48.0
Hentriacotane			5022			12444.6		47.6
Dotriacotane			4476			10389.8		50.8
Tritriacotane			2857			8294		40.6
Pentatriacotane			1399			6032.8		27.3
SS2					rage Response of Standards (from v			
Target Compounds	**IS Name**	**IS RESP**	**Cmpd Resp**		**Average Resp IS**	**Average Resp NS**		
Tetracosane	p-Terphenyl-d14	167718	8355		198028.8	22666.4	80.4	49.5
Pentacosane	Octacosane-d58	9841	5794		14460.8	20406.6		38.1
Hexacosane			5555			17674.6		42.2
Heptacosane			5129			18257.2		37.7
Octacosane			4360			16447.8		35.6
Nonacosane			4764			14558.6		44.0
Triacotane			3882			12149		42.9
Hentriacotane			3435			12444.6		37.1
Dotriacotane			2337			10389.8		30.2
Tritriacotane			2031			8294		32.9
Pentatriacotane			1707			6032.8		
SS3					rage Response of Standards (from v			
Target Compounds	**IS Name**	**IS RESP**	**Cmpd Resp**		**Average Resp IS**	**Average Resp NS**		
Tetracosane	p-Terphenyl-d14	178650	6513		198028.8	22666.4	78.9	51.7
Pentacosane	Octacosane-d58	10299	4694		14460.8	20406.6		41.4
Hexacosane			5897			17674.6		60.1
Heptacosane			5796			18257.2		57.1
Octacosane			5622			16447.8		61.5
Nonacosane			5297			14558.6		65.5
Triacotane			4414			12149		65.4
Hentriacotane			3837			12444.6		55.5
Dotriacotane			3264			10389.8		56.6
Tritriacotane			2509			8294		54.5
Pentatriacotane			1842			6032.8		
SS4					rage Response of Standards (from v			
Target Compounds	**IS Name**	**IS RESP**	**Cmpd Resp**		**Average Resp IS**	**Average Resp NS**		
Tetracosane	p-Terphenyl-d14	202711	9692		198028.8	22666.4	85.3	68.0
Pentacosane	Octacosane-d58	12624	6980		14460.8	20406.6		54.4
Hexacosane			6454			17674.6		58.0
Heptacosane			6800			18257.2		59.2
Octacosane			6297			16447.8		60.9
Nonacosane			5731			14558.6		62.6
Triacotane			4478			12149		58.6
Hentriacotane			4438			12444.6		56.7
Dotriacotane			3480			10389.8		53.2
Tritriacotane			2831			8294		54.3
Pentatriacotane			2153			6032.8		
SS5					rage Response of Standards (from v			
Target Compounds	**IS Name**	**IS RESP**	**Cmpd Resp**		**Average Resp IS**	**Average Resp NS**		
Tetracosane	p-Terphenyl-d14	165684	8566		198028.8	22666.4	84.3	83.9
Pentacosane	Octacosane-d58	10200	5061		14460.8	20406.6		55.0
Hexacosane			5547			17674.6		69.6
Heptacosane			5171			18257.2		62.9
Octacosane			4912			16447.8		66.3
Nonacosane			4830			14558.6		73.6
Triacotane			3697			12149		67.5
Hentriacotane			3388			12444.6		60.4
Dotriacotane			2744			10389.8		58.6
Tritriacotane			2482			8294		66.4
Pentatriacotane			1398			6032.8		

Table 18 GC/MS Analysis programme for alkane and PAH.

n-alkanes

	HP-5MS Column (30 m, 0.25 mm Diameter, 0.25 μm film thickness)
GC Conditions	
Injector Temperature (°C)	300
GC/MS Interface Temperature (°C)	300
Initial Oven Temperature (°C)	65
Initial Oven Hold Time (min)	5
Oven Temperature Ramp Rate 1 (°C/min)	10
Oven Temperature end 1 (°C)	250
Oven Hold Time 1 (min)	0
Oven Temperature Ramp Rate 2 (°C/min)	5
Final Oven Temperature (°C)	300
Final Oven Temperature Hold Time (min)	26.5
Carrier Gas	Helium
Carrier Gas Flow rate (ml/min)	1.0
Injection Mode	Splitless
MS Conditions	
Solvent Delay (min)	10
Data Collection Mode	SIM
Ion Monitored	57, 66, 71, 82, 85, 98
Dwell time	60 ms

PAH 1

	HP-5MS Column, (30 m, 0.25 mm Diameter, 0.25 μm film thickness)
GC Conditions	
Injector Temperature (°C)	300
GC/MS Interface Temperature (°C)	300
Initial Oven Temperature (°C)	65
Initial Oven Hold Time (min)	2
Oven Temperature Ramp Rate 1 (°C/min)	10
Oven Temperature end 1 (°C)	150
Oven Hold Time 1 (min)	0
Oven Temperature Ramp Rate 2 (°C/min)	4
Final Oven Temperature (°C)	300
Final Oven Temperature Hold Time (min)	30
Carrier Gas	Helium
Carrier Gas Flow rate (ml/min)	1.0
Injection Mode	Splitless
MS Conditions	
Solvent Delay (min)	10
Data Collection Mode	SIM
Ion Monitored (three groups)	1 (250, 252), 2 (274, 276, 278, 292), 3 (300)
Dwell time	1 (100 ms), 2 (80 ms), 3 (100 ms)

2.5 OC/EC Analysis-

A Sunset Laboratory Thermal-Optical Carbon Aerosol Analyzer was used to analyse for organic and elemental carbon concentration. The instrument uses thermal desorption in combination with optical transmission of laser light through the sample to speciate carbon collected on a quartz fibre filter (Sunset Laboratory Inc., 2000). 1cm2 punch from the quartz filter sample were used to analyse for the elemental, organic and total carbon using the EUSAAR2 (European Supersites for Atmospheric Aerosol Research) protocol for the measurement of carbon (Cavalli et al., 2010).

In a helium atmosphere, the temperature of the oven is increased to 700°C to remove the organic carbon from the sample which is converted to carbon dioxide as it passes to the manganese dioxide oxidizing oven. The carbon dioxide then mixes with hydrogen, over a heated nickel catalyst and it is converted to methane, which is measured using a flame ionization detector (FID). A second temperature ramp from 550 °C to 850 °C is then initialized with a helium/oxygen atmosphere in the oven. During this time the elemental carbon from the sample and pyrolysis products are oxidized and carried through the system and measured (Dall'Osto et al., 2011).

A fraction of collected organic carbon may be charred and pyrolized into EC during the initial heating process of the non oxidizing run and this could result in the report of less OC and more EC than actually present in a given sample. To correct for this problem, a tunable diode laser is used to determine the absorbance of the sample throughout the heating ramp cycle. The absorbance increases as OC is pyrolized to EC and decreases as EC and the pyrolized OC are desorbed during the second heating cycle. The point at which the laser absorbance returns to its initial value is considered the split point between OC and EC for quantification. Any carbon measured before the split is assigned as thermal OC, and any carbon measured after the split is

assigned as thermal EC(Bauer et al., 2009). Thermal EC and thermal OC are usually simply referred to as EC and OC.

The overall carbon response is based on a multi-point external calibration. The external methane standard is calibrated against an external multipoint calibration. The external methane standard is run at the end of every sample. This known amount is used to normalize the response factor for each sample. The software determines an initial FID response baseline prior to the desorption. The area at each point along the thermogram curve minus the baseline is multiplied against the calibration response to determine the carbon. The data are summed over the range to yield the total carbon results.

Instrument calibration was done with solutions of sucrose standard which were prepared (4.20 µg/µl, 0.42 µg/µl and 0.21 µg/µl). 1 cm^2 of clean unexposed filter paper was punched using a puncher on a clean aluminium foil surface and pinched loose with a paper clip. 10 µl of the standard was dropped on the filter cut out and allowed to dry for about30 minutes. Then the instrument was used and run using the operating procedure for the "SUNSET THERMAL-OPTICAL CARBON ANALYSER" instrument should be followed -

The powers for both instruments and computer were switched on and then the gases (Air, Helium, Helium/Oxygen, Hydrogen and Helium/Methane) were switched on and gas pressures set.

The programme for the carbon analyser was started on the computer and it was ensured that gas flows were set within the expected ranges and hydrogen was set last. The machine was then allowed to stand for about 30 minutes.

A file from the PAR folder was selected and a name entered for the raw data file, the sample ID and analyst names and size of punch area (1 cm^2 in my case.)

The instrument was then allowed to be ready for running (indicated by the green bar at the bottom of the screen which shows ‘safe to put a new sample’). To place the sample the quartz door to the front of the oven was opened and the punched out sample, for analysis, was placed on the boat of the OC/EC spoon using twisters, and replaced in by sliding the spoon gently until it stopped. This was followed by the door being closed and tightened with the metal clip followed by clicking the “start analysis” button for the analysis of the sample.

This procedure was repeated for the 3 concentrations to ensure the instrument is working efficiently.

When the instrument was calibrated and found to be working accurately, blank and filter samples were then run where 1 cm^2 of the filter paper to be analysed were punched using the puncher on a clean aluminium foil surface and pinched loose with a paper clip.

For the analysis of samples stored in the freezer the samples were brought out in the stored wrapping (plastic bags and foil wrapping) and allowed to stand for some time to allow the filter to reach room temperature. The foil paper casing was carefully opened to prevent exposure of filter to moisture.

The instrument was turned on and used as described above in the explanation of the SOP for the “SUNSET THERMAL-OPTICAL CARBON ANALYSER”.

CHAPTER 3- Cooking Source profile.

This chapter presents concentration of particulate matter collected from cooking source in a controlled environment. The aim of the sampling was to characterize the emissions from various cooking styles and compare them among themselves as well as compare them with the limited existing profiles available.

This chapter contains some sections of verbatim text adapted from the following review article published as part of this PhD:

Abdullahi, L, Delgado Saborit, JM & Harrison, RM 2013, 'Emissions and indoor concentrations of particulate matter and its specific chemical components from cooking: A review' **Atmospheric Environment**, vol 71, pp. 260- 294.

3.1 Introduction

With the identification that food cooking is one of the major sources of pollution in the indoor environment as well it being an important source of the fine organic aerosol in urban environments, a better understanding of what is emitted during the process is important.

Some studies have characterised emissions from cooking in both controlled environments , involving cooking experiments, and also in real-world residential and commercial kitchens where measurements were taken (Li et al., 2003, Rogge et al., 1993, He et al., 2004c, Schauer et al., 1999a, Lee et al., 2001b, Robinson et al., 2006, Rogge et al., 1991). When sampling in controlled experimental setups it is assumed that the measurements are influenced mainly by the fuel used and the food being cooked while in actual real life kitchens measurement of emissions are influenced by many factors such as room arrangement, building materials, outdoor infiltration, other combustion devices, ventilation, and cooking methods(Huboyo et al., 2011) . There is a need for both types of microenvironments so as to have a neutral representation of what actually is emitted from the food and also have a representation of what the population is actually being exposed to in their daily lives

In the investigations by Rogge et al., 1991, Robinson et al., 2006 and He et al., 2004 they were mainly focused on the determination of semi-volatile organic compounds as well as the

speciation of organic aerosols released from the cooking being carried out and in the end Robinson et al., 2006 strongly suggested the use of organic molecular organic tracers such as *n*-hexadecanoic (palmitic) acid, *n*-octadecanoic (stearic) acid, 9-hexadecenoic (palmitoleic) acid, 9-octadecenoic (oleic) acid, and cholesterol as source contribution estimates for food cooking. However these studies were limited to only specific cooking styles as well as they were carried out in different parts of the world : Rogge et al., 1993a in Los Angeles, He et al., 2004 in China, Schauer et al., 1999, Lee et al., 2001, Robinson et al., 2006 Pittsburgh, Pennsylvania, Li et al 2003 in Southern Taiwan.

Different cooking methods and fuels are used for the preparation of food around the world which affect the particle emissions as well as the physical and chemical properties of the particles generated (See and Balasubramanian, 2008, Lee et al., 2001b). $PM_{2.5}$ concentration were found to be low , medium and high when the cooking method were predominantly steaming , boiling and frying respectively by Lee et al., 2001 where samples were taken in a Korean barbeque restaurant(frying), a Chinese hot pot restaurant (boiling) and a Chinese dim sum restaurant (steaming). See and Balasubramanian, 2008 similarly found that deep-frying gave rise to the largest amount of $PM_{2.5}$ and most chemical components, followed by pan-frying, stir-frying, boiling, and steaming. As such the concentration of cooking can vary based on these parameters such as cooking method and ingredients and may as well be unique for every region of the world.

He et al also collected samples from two Chinese restaurants with predominately two cooking styles; Hunan and Cantonese cooking (He et al., 2004). The samples were similarly collected through the overhead exhaust hood at the exit of the exhaust duct. The samples were taken at lunchtime and supper in the evening for each restaurant. The Cantonese Style involved mainly frying, stewing or braising of food. Schauer et al., 2001 collected samples when cooking was carried out using a large institutional-scale deep fryer and a large industrial-scale electric grill

where commercially distributed food products were being prepared. Soya beans and canola oil were used to stir fry vegetables while hydrogenated soybean oil were used to deep fry chips. Samples were collected downstream from the grease extractors located in the ventilation system above the appliances (Schauer et al., 2001).

None of the reported research so far was conducted in the UK and none involved the experiments on various cooking styles being carried out by the same researcher in the same location over the same period.

The aims of this chapter are

- Characterization of the chemical composition of PM from cooking emissions.
- Preparation of cooking source profiles for various cooking styles.

3.2 Sampling and analytical methods.

Samples were taken from the laboratory trailer kitchen (a controlled environment) described in Chapter 2 and the filters collected were analysed using methods described in chapter 2.

The quartz filter samples were used to analyse OC and EC using the carbon analyser and GC-MS was used to analyse for PAHs, alkanes, Acids, Hopanes, Sterols and glycerides.

The PTFE filter samples were used to obtain gravimetric concentrations of PM.

The following sections include the analysis of the results obtained from the laboratory analysis of samples collected. These data are compared with data obtained from previous studies on cooking emissions in Table 19, which involved similar sampling techniques and analysis. For instance Zhao et al., 2007c collected samples from commercial restaurant using two medium-volume samplers, located on the roof, to collect $PM_{2.5}$ on quartz fibre filters. The samples were collected directly from the exit of exhaust during the periods of lunch and supper for 2 hours for each sample. In this study the Western-style fast food used frying

techniques and beef and chicken were the main ingredient while at the Chinese restaurants the main ingredients for cooking were pork, poultry, beef, seafood, vegetables.

Differences in cooking time for each cooking style generally existed ranging from 40 mins for western cooking to 1 hour for Chinese, Indian and African cooking but the time differences were about the same (range of 45 mins to 60 mins) so an assumption was made that the differences in time was not significant considering that calculation will be made based on the sample times to obtain the sample volume.

Table 19 Studies on emissions from cooking

STUDY	SAMPLING CONDITIONS
Schauer et al., 1999a Characterise organic compound composition emitted during meat charbroiling **Schauer et al., 2002** Characterise organic compound composition emitted during oil cooking	Emissions sampled in the ventilation system of a commercial kitchen downstream of the filter and grease extractor. Sampling time was 85 min. Dilution tunnel: mix exhaust emissions with 25- to 180-fold excess of HEPA filtered air. 1.8 µm AIHL-design cyclone separators upstream of samplers. Flow rate in each sampling train was 10 L/min, except sampling train a) at 30 L/min and sampling train g) at 0.2 L/min. Organic compounds collected using: a) 1 XAD coated denuder upstream of 3 quartz filters in parallel followed by 2 PUFs in series. b) 3 quartz filters followed each by 2 PUFs in series. EC/OC collected using: c) 2 quartz filters in series Mass emissions, trace metals and organic acids collected using: d) Teflon filter upstream of two KOH impregnated quartz fibre filters Mass emissions & soluble ions collected using: e) Teflon filter VOC collected using: f) 6-L SUMA canister downstream of teflon filter e) Carbonyls collected using: g) DNPH-impregnated C18 cartridges
Svendsen et al., 2002 Characterise aldehydes and fat aerosol collected in the breathing zone of the cook in fumes from commercial restaurants.	Personal exposure sampler with inlets located in the shoulder of the cook of 19 commercial kitchens using deep frying devices equipped with ventilation hoods. Aldehydes were collected a sampling device containing silica impregnated with 2,4-dinitrophenyl hydrazine. Flow rate was 1.5 L/min during 1.5-2.5 hours. Fat aerosol collected onto pre-weighted one glass fibre filter (Nucleopore AAA). Flow rate, 2 L/min during 65 to 200 mL. Total number concentration was measured with TSI 3936 SMPS used to measure the PAHs were collected onto glass fibre filters in a filter holder and 2 XAD-2 tubes downstream. Flow rate, 1 L/min during 200 min.
McDonald et al., 2003 Characterise organic compound emission composition emitted during charbroiling and grilling of chicken and beef	University lab kitchen following commercial standard procedures. Emissions collected at the end of a residence chamber to allow the gas/particle equilibrium. 2.5 µm cyclone separators upstream of samplers. Flow rate in each sampling train was 113 L/min. Samples collected on Teflon filter for $PM_{2.5}$ and elements. Samples collected on quartz filters for carbon and ion analysis Samples collected on Teflon-impregnated glass fibre (TIGF) filter followed by a PUF/XAD-4/PUF sandwich cartridge for speciated particle-phase and semi-VOCs. CO was measured using non-dispersive infrared analyser.
Zhu and Wang, 2003 Characterise PAH emitted in commercial and domestic Chinese kitchens	A sampler was located in a new kitchen 0.5 m from the pan (cooking methods) and in the centre of the kitchen (domestic and commercial kitchens). In all cases, the sampler was 1.5 m above the ground level. All doors and windows were closed during cooking. Electric hobs were used for cooking. Samples were collected over 100 mins to test different cooking methods, and over 2 consecutive days for 12-h (8:00 – 20:00) in domestic and commercial kitchens. Low noise small samplers (MP-15CF) operated at 1.0 l/min equipped with a Whatman glass filter (GFF, 25 mm) collected particle bound PAHs and a XAD-2.5 g cartridge collected the gaseous PAHs.
Chen and Chen, 2003 Characterise PAHs in fumes during frying of chicken.	Emissions collected on adsorption wool fitted on the cover of frying tank (closely tight during sampling)
Li et al., 2003 Characterise PAHs in fumes during cooking of different styles	Emissions collected isokinetically from the exhaust vent in commercial kitchens. Three consecutive samples were collected at 10L/min for 45 min during the cooking time. Particle bound PAHs were collected on a tube-type glass fibre thimble (25x90 mm). Gaseous PAHs were collected onto a 5-cm polyurethane foam (PUF) followed by a 2.5 cm Xad-16 resin cartridge supported by a 2.5 cm PUF.
He et al., 2004b Characterise fumes emitted during Chinese style cooking	Samples collected at the exit of the exhaust vent of two commercial kitchens. Sampling times were 90-120min at lunchtime and dinner. Samples collected onto two honeycomb sampler and a three stage cascade impactor to collect $PM_{2.5}$ at 25 L/min.

	One honeycomb contained PTFE filters for particle mass determination and and ionic species analysis. The second honeycomb and the cascade impactor were loaded with quartz filters (Pallflex 2500QAT-UP) for the determination of EC/OC and organic speciation.
He et al., 2004c Characterise fumes emitted during Chinese style cooking	Samples collected at 40-60 cm at leeway from the exhaust vent of two commercial kitchens. Sampling times were 100-120min at lunchtime, and 45 minutes at dinner. Samples collected onto quartz fibre filters with a three stage cascade impactor (<10um, 10-2.5 um and <2.5 um) at 25 L/min.
See et al., 2006; See and Balasubrabramanian, 2006b Characterise PAH and metal composition emitted during Chinese, Malay and Indian style commercial cooking **See and Balasubrabramanian, 2006a, 2008** Characterise emissions from 5 types of cooking methods (steaming, boiling, stir-frying, pan-frying and deep- frying)	Sample collected at 1.5m above ground level at the opposite site of a 4 LPG burners stove in commercial food stalls.(See et al., 2006; See and Balasubrabramanian, 2006b) Sample collected at 1.5m above ground level and 0.2 m from a 2-burner domestic stove with no ventilation. Samples collected during cooking activities. (See and Balasubrabramanian, 2006a, 2008). Samples collected for 12 hours during cooking and non-cooking activities. A MiniVol portable air sampler (Airmetrics) collected $PM_{2.5}$ at a flow rate of 5 L/min onto: - 47mm 2 µm PTFE Teflon filter for gravimetric, metals and ion analysis. - 47mm QMA quartz filters for PAH
Zhao et al., 2007a, b Characterise organic compound emission composition emitted during Chinese and Western style cooking	Emissions sampled at the exhaust vent of the ventilation system of commercial kitchens downstream of the filter treatment methods. Samples collected during rush hour at lunch and dinner times. Sampling time was 120 min. 2 medium-volume samplers at a flow rate of 78 L/min collected samples in 90mm quartz fibre filter. 2 Dustraks (TSI) monitored the relative concentrations of $PM_{2.5}$ and PM_{10}. Background $PM_{2.5}$ was collected in the city using a hi-volume sampler (Andersen).
Sjaastad and Svendsen, 2008; Sjaastad et al., 2010; Sjaastad and Svendsen, 2009 Characterise PAHs, aldehydes and particulate matter collected in the breathing zone of the cook in fumes from frying a beefsteak.	Model kitchen (19 m2) containing gas or electric hobs and a canopy fume hood. Personal exposure sampler with inlets located in the shoulder of the cook. PAHs were collected onto glass fibre filters in a filter holder and 2 XAD-2 tubes downstream. Flow rate, 1 L/min during 200 min. Aldehydes were collected into stainless steel sorbent tubes filled with 220 mg Tenax TA. Flow rate, 100 mL/min for 10-200 min. Total particles collected onto pre-weighted double Gelman glass fibre filters. Flow rate, 2 L/min during 65 to 200 mL. Total number concentration was measured with TSI 3936 SMPS.
Hildemann et al., 1991a	Commercial scale kitchen -Sampling port located above the cooking surface, below the extractor fan.
Li et al., 1993	Domestic kitchen with a gas stove Sampling ports 3m away from the gas stove
Abt et al., 2000a, b	Domestic kitchen with gas and electric stoves. Samples collected over 6-day periods Equipment located in an indoor location adjacent to the kitchen.
Dennekamp et al., 2001	Laboratory kitchen with gas and electric stoves Sampling inlet at face level in front of the cooker
Wallace et al., 2004	Domestic kitchen using gas stove Measurements performed in the duct of the ventilation system.
Wallace et al., 2006	Personal and indoor (living room) measurements for 7 days in free-style living conditions.
Hussein et al., 2006	Domestic kitchen using an electrical stove and adjacent living room.Continuous measurement for 15 days at 3 min intervals Sampling ports at 1.5m from the ground and 1m (kitchen) and 5m (adjacent room) from the stove.
Yeung and To, 2008	Laboratory kitchen (168m3) with gas stove and electric griddle. Fume hood installed above cooking area.
Buonanno et al., 2009 **Buonanno et al., 2011**	Open plan laboratory kitchen (80m2) using gas and electrical stoves. Sampling 2 meters away from the stove for 8-10 mins.
Buonanno et al., 2010	15 pizzerias Sampling 2 meters away from the stove for 8-10 mins.
Glytsos et al., 2010	Laboratory room (60m3) Electric stove Sampling ports 0.9 m above the floor.

3.3 Gravimetric concentration.

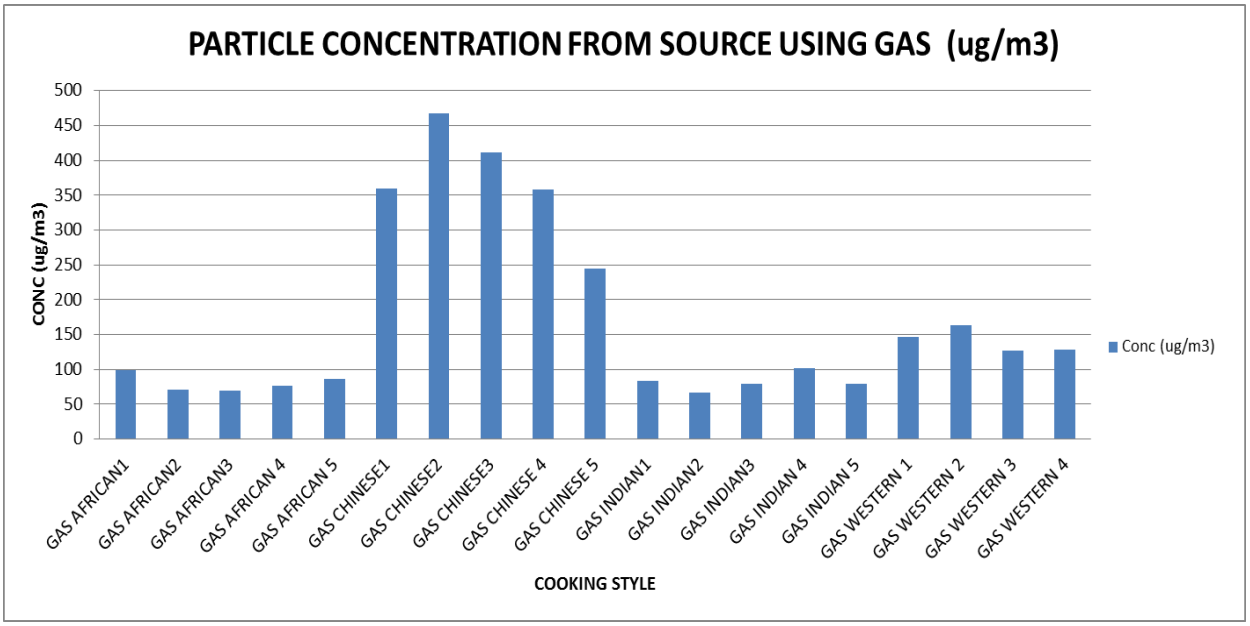


Figure 21 Particulate matter Concentration at cooking source using gas (µg/m^3)

High concentrations of PM are observed in Figure 21 for Chinese cooking with a range of between 244 µg/m^3 and 467 µg/m^3. Indian style cooking is found to release less PM concentration of all the cooking styles with concentrations as low as 67 ug/m3 in Table 20 and Figure 22; See et al 2006 had similar concentration at their sampling point in a commercial food stall where Chinese food was being made. They found that PM concentration was 312.4 ug/m^3 which lies within the range of concentration obtained in this study. He et al measured slightly higher concentrations of PM in China 1406.3 ug/m^3 and 672 ug/m^3 at exits of exhaust duct of Hunan and Cantonese restaurants respectively. The process of stir frying involved in Chinese cooking leads to oil, meat and other ingredients to be able to reach very high temperatures leading to high breakdown of these food resulting in large amount of particle generated compared to other cooking styles.

The next cooking method to release higher concentration of PM is the western style cooking which like the Chinese cooking is based on frying in oil.

Similar concentrations were observed for Indian and African and cooking which both involved some frying, stewing and boiling. Indian cooking emitted a range of 67-102 ug/m^3 while African cooking had concentration between 70-99 ug/m^3.

Cooking with oil is likely to generate more particles than boiling of water and this is attributed to the higher temperature needed to boil oil as against water; for instance corn oil has a boiling point of 245°C while water has a boiling point of 100°C (Sjaastad, 2010). This physical property enables the oil droplets generated during cooking to exist as particles as against the less volatile water droplets (Sjaastad, 2010). As compared to oil cooking, the boiling of water leads to the generation of steam resulting in higher humidity in the kitchen resulting in the hydroscopic growth of particles (Wallace and Howard-Reed 2002) and water vapour condensing on UFP forming larger particles (See and Balasubramanian 2006a; Sjaastad, 2010).

The one-way analysis of variance (ANOVA) is used to determine whether there are any statistically significant differences between the means of the various PM concentrations for the different cooking styles. The results showed that there was significant difference between all the means sig (0.001)

Table 20 Concentration of PM emitted from cooking source using gas (µg/m^3)

	AVERAGE	STD DEV
AFRICAN	80.6	12.3
CHINESE	367.8	82.5
INDIAN	82.4	12.7
WESTERN	141.3	17.2

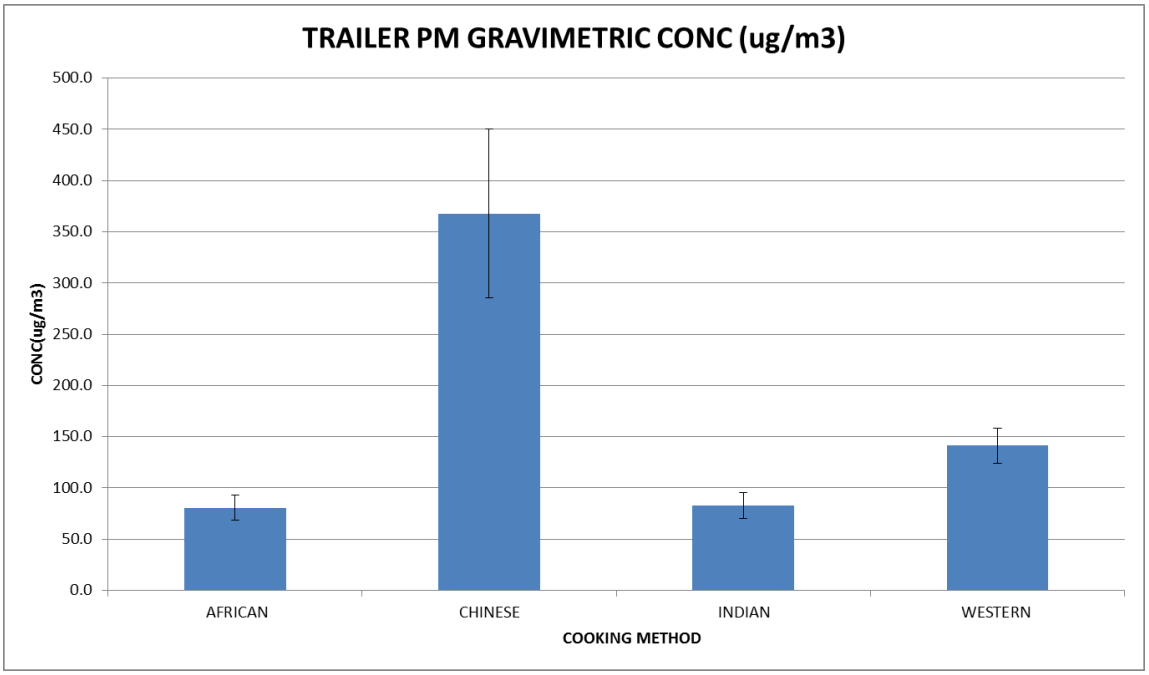


Figure 22 figure showing concentration of PM emitted from cooking source using gas (with error bars)

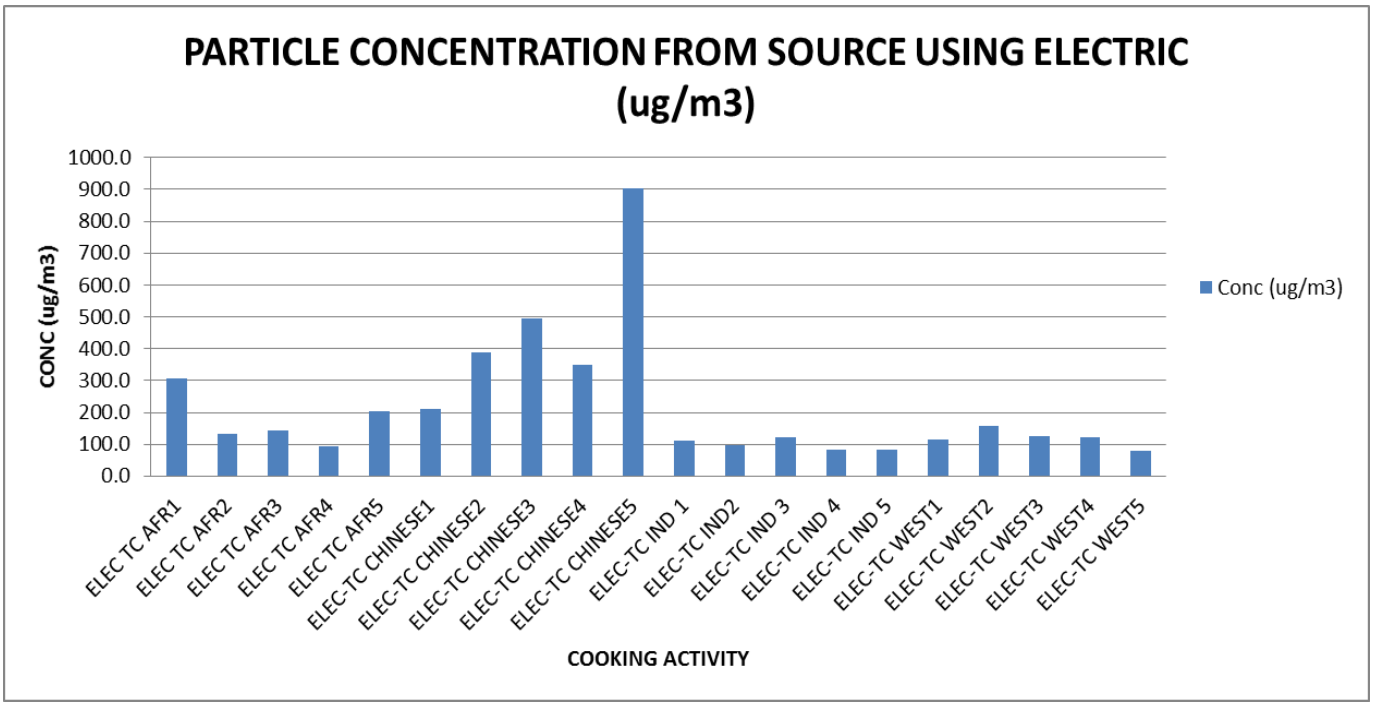


Figure 23 Particulate matter Concentration at cooking source cooking with electric (µg/m^3)

Figure 23 show the PM concentration at the trailer (TC-TRAILER COOKING) for the various cooking styles. Generally the concentrations generated are slightly higher during cooking with electric with a range of 80-900µg/m^3 compared to the range of 60-460µg/m^3 gotten when cooking with gas.

Table 21 Concentration of PM emitted from cooking source using electric ($\mu g/m^3$)

	AVERAGE	STD CEV
ELEC TC AFR	176.6	83.4
ELEC-TC CHINESE	469.3	263.4
ELEC-TC IND	99.3	18.6
ELEC-TC WEST	119.7	27.8

When Table 21 and Table 20 are compared it is seen that the Chinese cooking style emits the highest concentration of PM. African and western style cooking

3.4 Concentration of compound emitted from various cooking styles.

Table 22,Table 23,Table 24, **Figure 24** and **Figure 25** are tables and figures that present the concentrations of organic compounds that were obtained from the analysis of filters collected in the trailer kitchen.

Alkanes

In this study higher concentrations of heptacosane were observed at the cooking source for Indian cooking with low concentration of tetratriacontane 2.71 $\mu g/m^3$ and 0.18 $\mu g/m^3$ respectively. This was the trend observed for all the cooking types with African generally emitting less concentrations (heptacosane 0.41 $\mu g/m^3$ and tetratriacotane 0.07 $\mu g/m^3$), highest concentrations were observed in Indian cooking followed by western style cooking then Chinese cooking. A very high concentration of 2.88$\mu g/m^3$ was observed for tritriacotane in Chinese cooking, with Indian African and Western style cooking emitting 0.89 $\mu g/m^3$,0.4 $\mu g/m^3$,1.11 $\mu g/m^3$ of the same alkane.

Similarly in previous studies the distribution of n-alkanes emitted from Chinese restaurants have generally been observed to be substantially different from the distribution from meat cooking (Rogge et al., 1991; Schauer et al., 1999a; He et al., 2004b) and similar to alkane patterns from

frying vegetables in seed oils (Schauer et al., 1999a; Schauer et al., 2002). Emission of n-alkanes from cooking consisted of a negligible fraction of the total quantified organic mass emitted and is dependent on the cooking conditions (Rogge et al., 1991; He at al., 2004b)as seen in Table 26. Hildemann et al., (1991a) reported that the n-alkane concentration release rate increased from frying to charbroiling of meat with extra lean meat releasing less compounds than regular meat (Hildemann et al., 1991a). This was similar to observations by Rogge et al., (1991), where charbroiling was found to produce three times the mass of n-alkanes than frying of meat (16 mg/kg of charbroiling meat as against 5.5 mg/kg of frying meat). Rogge et al. (1991) also observed that charbroiling regular meat released four times the mass compared to extra lean meat (thus affected by fat content of meat). In this study the n alkane concentration for Indian and African cooking are found to be a more significant fraction of the total organic mass as seen in Table 25.

Similar to previous studies by Zhao et al in 2007 where Western style fast food cooking had been observed to emit double the concentration of n-alkanes per mg particulate organic matter (POM) compared to Chinese cooking, concentration of alkanes is less in Chinese style cooking in this study. The n-alkanes have a C_{max} at Pentacosane(C_{25}) for western fast food (Zhao et al., 2007a) and meat cooking (Rogge et al., 1991). Chinese cooking exhibits a C_{max} at Nonacosane or Hentriacontane (C_{29} or C_{31}) taken as an indication of the presence of vegetables during cooking operations. In this study Indian cooking had a C_{max} at Heptacosane(C_{27}), Western at Hexacosane(C_{26}), African at Nonacosane(C_{29}) and Chinese at Tritriacotane(C_{33}).

PAH

During Chinese and Indian cooking highest concentration of PAH was for dibenz(ah)anthacene 1.96 µg/m^3 and 0.96 µg/m^3respectively. For western cooking highest concentration were found for benzo(b)fluoranthene 1.50 µg/m^3. Generally African food was found to release lower concentrations of PAH than the other cooking styles.

A similar trend was observed when Chinese cooking and Indian cooking were compared: higher PAH concentrations were observed for Chinese cooking due to stir frying and higher cooking temperature, whilst the Indian cooking style generated the lower PAH concentrations. Indian cooking emitted large amounts of volatile PAH with lower molecular weight like naphthalene, fluoranthene and phenanthrene attributed to low temperature cooking, such as simmering (See et al., 2006). Chinese cooking, on the other hand, was found to emit higher molecular weight PAHs such as benzo[b]fluoranthene, indeno[1,2,3-cd]pyrene and benzo[g,h,i]perylene. These trends were attributed to the cooking methods employed in each type of cooking from the amount of food cooked, the amount and type of oil used, to the temperatures reached during cooking, and cooking time (See et al., 2006).

The effect of the cooking method was also examined by See and Balasubramanian (2008), who found that techniques that involve the use of oil at high temperatures, such as stir frying, pan-frying and deep-frying, released higher amount of PAH compared with those that involve the use of water, such as boiling and steaming. This is consistent with work of Schauer et al. (2002). Higher quantities of oil are generally used in stir frying, commonly used in Malay and Chinese cooking, than simmering which is the most common technique used for preparation of Indian dishes. In addition, high temperature frying was found to lead to production of higher molecular weight PAHs, while low temperature cooking results in formation of more low molecular weight PAHs (See et al., 2006). McDonald et al. (2003) compared the PAH emissions from charbroiling and grilling meat and found that PAH emissions from charbroiling were about 3–5 times more than when food was grilled. This was attributed to the contact of the lipid material dripping from the meat (during cooking) onto the cooking appliance. Thus, the higher PAH concentrations observed during charbroiling were due to the direct access of lipids onto the hot flame compared to the cooler griddle surface used in grilling (McDonald et al., 2003).

The emission of PAHs in cooking fumes, not only is related to the cooking method, but also to the cooking ingredients. Schauer et al. (1999a; 2002) studied the emissions of cooking fumes for

charbroiling hamburger meat (1999a) and frying vegetables (2002). They found that cooking meat produced far greater PAH concentrations than frying vegetables. Zhu and Wang (2003) studied the emissions of low and high fat food using different cooking methods. The frying of low fat foods was observed to lead to the generation of more PAH than the broiling. This was not the case for high fat food which exhibited the reverse with higher concentration of PAH detected when the food was broiled (Zhu and Wang, 2003). This was illustrated when low fat fish produced a higher level of PAH when fried than when broiled, and pork chops produced higher PAH when broiled than when fried.

Acid

Higher acid concentrations were observed in Chinese cooking with 9-Octadecenoic acid being the acid with highest concentration for this style of cooking (6.49 $\mu g/m^3$). High concentrations of hexadecanoic acid were also observed in Chinese and all other cooking styles with concentration of 4.22 $\mu g/m^3$, 2.03 $\mu g/m^3$,1.23 $\mu g/m^3$ and 0.84 $\mu g/m^3$ for Chinese, African, western and Indian cooking respectively.

Due to the fact that meat and oils used in cooking contain fats made up of saturated and unsaturated fatty acid esters of glycerol, chemical processes that typically occur during high temperature treatment of food are the degradation of sugars, pyrolysis of proteins and amino acids and the degradation of fats (Svendsen et al., 2002). The cooking process leads to production of free fatty acids, free glycerol and mono- and diglycerides (Nolte et al., 1999).

In previous studies by Zhao et al ,Western fast food cooking found that the quantified saturated fatty acids observed a range from C_6 to C_{20} with distinct even to odd carbon preference and a predominance of palmitic acid (Zhao et al., 2007a). Chinese cooking was found to emit C_6-C_{24} fatty acids with a similar even to odd carbon preference and palmitic acid preference similar to meat cooking (Rogge et al., 1991; He et al., 2004b) and seed oil cooking (Schauer et al., 2002). The most common unsaturated fatty acids observed were oleic acid and linoleic acid for Chinese cooking

(Zhao et al., 2007b; He et al., 2004b). The most prominent organic compound released from American cooking is oleic acid (Rogge et al., 1991; Schauer et al., 1999a; Schauer et al., 2002; He et al., 2004b).

The concentration of emitted saturated fatty acids in Western fast food was found to be 13 times higher than in Chinese cooking while unsaturated fatty acid concentrations were only two times higher, attributed to ingredients and cooking temperature. High concentrations of nonanoic acid emissions are observed in both Chinese and Western style fast food cooking with a higher ratio of nonanoic acid to other acids (C_8-C_{10}) in Western style fast food. Schauer et al. (1999a; Schauer et al., 2002) compared the emissions of fatty acids from different ingredients, such as meat and vegetables. They found that charbroiling hamburger meat released more fatty acids than frying vegetables. They also found that stir frying released more fatty acids than deep frying.

Aldehydes

A recent IARC monograph reported that cooking, in particular frying, generates substantial amounts of certain gaseous pollutants such as formaldehyde (IARC, 2006), acetaldehyde (IARC, 1999), acrylamide (IARC, 1994) and acrolein (IARC, 1995). These compounds have not been studied in this study and so no comparism can be done at this point. However hereunder describes the main results found in the literature about aldehydes.

Concentration distributions have been found to be similar for Western and Chinese style cooking for most of the aldehydes, except for nonanal, which is one order of magnitude higher in Western cooking (Zhao et al., 2007b).

Similar to what was observed for other organic species, the type of ingredient cooked is also key in the release of aldehyde emissions during cooking. The studies of Schauer et al. (1999a; Schauer et al., 2002) show that charbroiling hamburger meat emits more aldehydes than frying vegetables.

Sjaastad et al. (2010) used a model kitchen similar to a Western European restaurant to assess if higher mutagenic aldehydes were emitted during the frying of beefsteak on an electric or gas stove with margarine or soya bean oil as the frying fat oil. It was found that mutagenic aldehydes were detected in the breathing zone of cooks in the range of non-detectable to 61.80 µg/m^3 (Sjaastad et al., 2010). They also found that higher exposures to these components were more pronounced when frying on a gas stove instead of an electric stove which may cause adverse health effects especially for people occupationally exposed to these fumes (Sjaastad et al., 2010). An earlier study of Sjaastad and Svendsen (2008) had involved the frying of beef steak using margarine, rapeseed oil, soybean oil or virgin olive oil as frying fat in similar conditions as a regular Norwegian home (in terms of ventilation conditions and frying procedure).They recorded mutagenic aldehyde concentrations ranging from non-detectable to 25.33 µg/m^3 (Sjaastad and Svendsen, 2008). They also observed statistically significantly higher levels of mutagenic aldehydes and particles when margarine was used as the cooking fat compared to the other oil.

Generally it was found that there was a shortage of literature on characterisation of emissions from cooking using electric stove with most studies reporting mainly particulate matter mass and not the organic composition. In this study, a look at the emissions of compounds emitted at the trailer kitchen(cooking source) using an electric hob was made and the concentrations were reported in Table 29, Table 30, Table 31 and Table 32 for the various cooking styles. It was observed that compared to the gas cooking concentrations were higher than when electric hob was used, for instance the average concentration of glycerides cooking Indian food was 0.8µg/m^3 when cooking with gas as against 0.45µg/m^3 when using electric. Acid concentrations also showed a similar trend with average concentration of the group of acids of 0.42µg/m^3 total emitted from gas cooking against 0.2µg/m^3 when electric source of heat was used. For Chinese cooking 1.35µg/$m^3$of total acid as compared to 0.24µg/m^3 emitted when electric was used.

Similar to cooking with gas it was observed that emission during Chinese cooking produced higher concentration of compounds, followed by western style cooking then Indian and finally African.

Total concentration for the various cooking methods for glyceride, sterol and acid respectively were found to be; Chinese (0.8, 0.3 and 1µg/m^3), western (0.7,0.3 and 0.2µg/m^3) Indian (0.5,0.2 and 0.2µg/m^3) and African(0.2,0.1 and 0.8µg/m^3) showing the trend of total concentration.

Table 22 Chemical composition of PM emitted from cooking source µg/m^3 using gas

ALKANES	INDIAN	WESTERN	AFRICAN	CHINESE
Tetracosane	0.67	0.50	0.63	1.28
Pentacosane	0.90	0.62	0.41	0.66
Hexacosane	1.40	1.93	0.49	0.75
Heptacosane	2.71	1.78	0.41	0.87
Octacosane	1.09	1.03	0.38	0.90
Nonacosane	1.22	1.31	0.59	1.33
Triacotane	0.98	0.96	0.35	0.99
Hentriacotane	0.92	0.88	0.34	1.30
Dotriacotane	0.81	0.77	0.30	0.94
Tritriacotane	0.89	1.11	0.40	2.88
Tetratriacotane	0.18	0.16	0.07	0.18
Pentatriacotane	0.64	0.61	0.29	0.90
PAH				
Benzo[b]fluoranthene	0.54	1.50	0.27	0.90
Benzo[k]fluoranthene	0.32	0.11	0.11	0.35
Benzo[e]pyrene	0.07	0.10	0.14	0.67
Benzo[a]pyrene	0.85	0.97	0.38	1.47
Perylene	0.61	0.76	0.20	1.70
Indeno[123-cd]pyrene	0.68	1.37	0.32	0.97
Dibenz[ah]anthracene	0.96	1.44	0.44	1.96
Picene	0.42	0.68	0.48	0.67
Benzo[ghi]perylene	0.75	1.28	0.23	1.66
Coronene	0.14	1.09	0.33	2.39
1-Monomyristin	0.93	2.64	0.29	2.25
1-Monopalmitin	0.70	2.90	0.24	3.84
1-Monoolein	0.80	3.23	0.53	3.37
1-Monostearin	0.95	1.56	0.42	2.05
Levoglucosan	1.04	0.78	0.31	1.18
Cholesterol	0.14	0.16	0.06	0.15
undecanoic	0.24	1.47	0.13	0.77
octanedioic	0.14	0.83	0.20	1.63
dodecanoic	0.54	0.52	0.26	0.83
nonanedioic	0.13	0.18	0.46	0.58
tridecanoic	0.10	0.45	0.02	0.11
tetradecanoic	0.19	0.67	0.10	0.30
pentadecanoic	0.45	0.42	0.15	0.45

Hexadecanoic	0.84	1.23	2.03	4.22
heptadecanoic	0.01	0.04	0.01	0.03
9,12-Octadecadienoic	0.95	1.11	0.97	4.11
9-Octadecenoic	2.32	2.24	1.96	6.49
Octadecanoic	0.38	0.23	0.48	1.81
nonadecanoic	0.02	0.02	0.01	0.04
eicosanoic	0.02	0.02	0.01	0.06
docosanoic	0.28	0.38	0.02	0.09
tetracosanoic	0.05	0.07	0.02	0.08

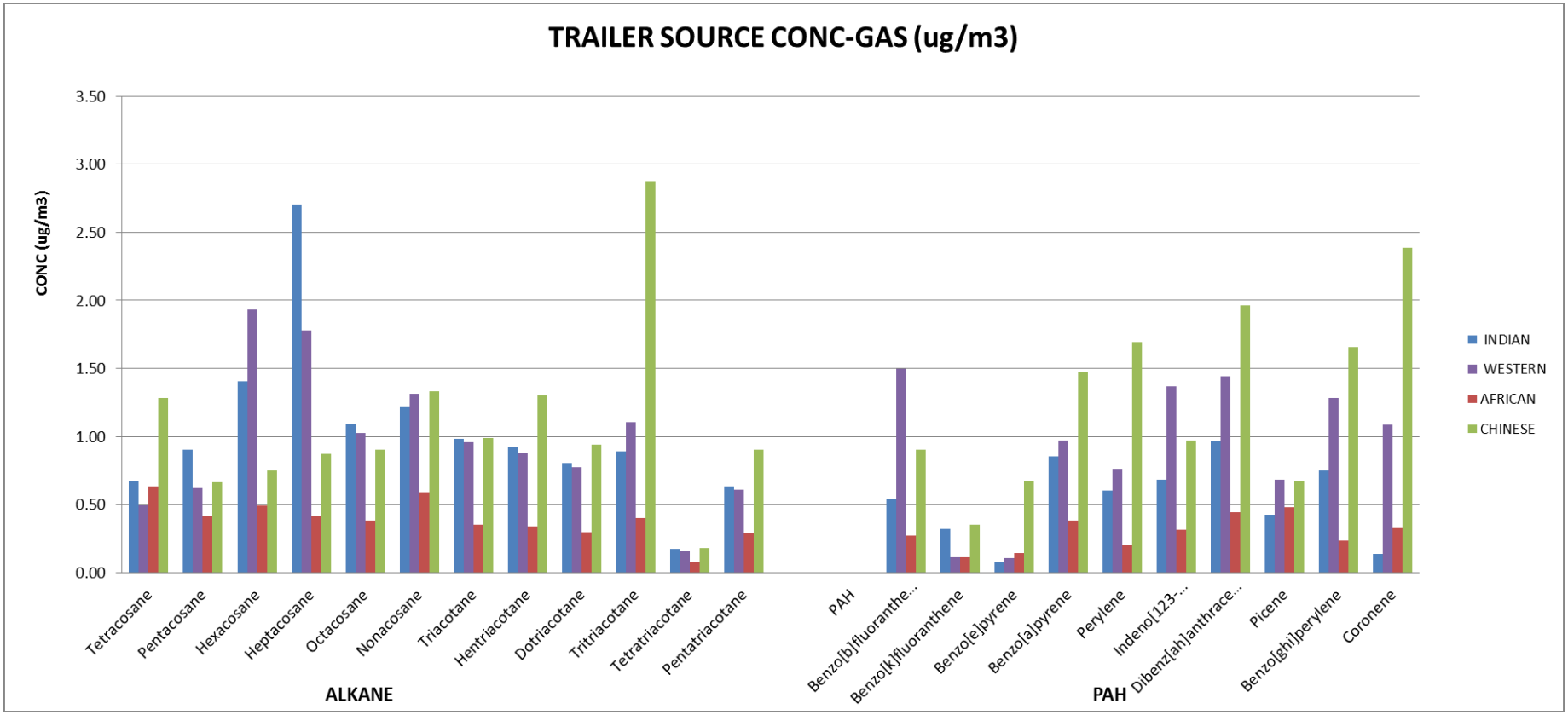


Figure 24 Concentration of compound (Alkane and PAH) emitted at cooking source (µg/m^3)

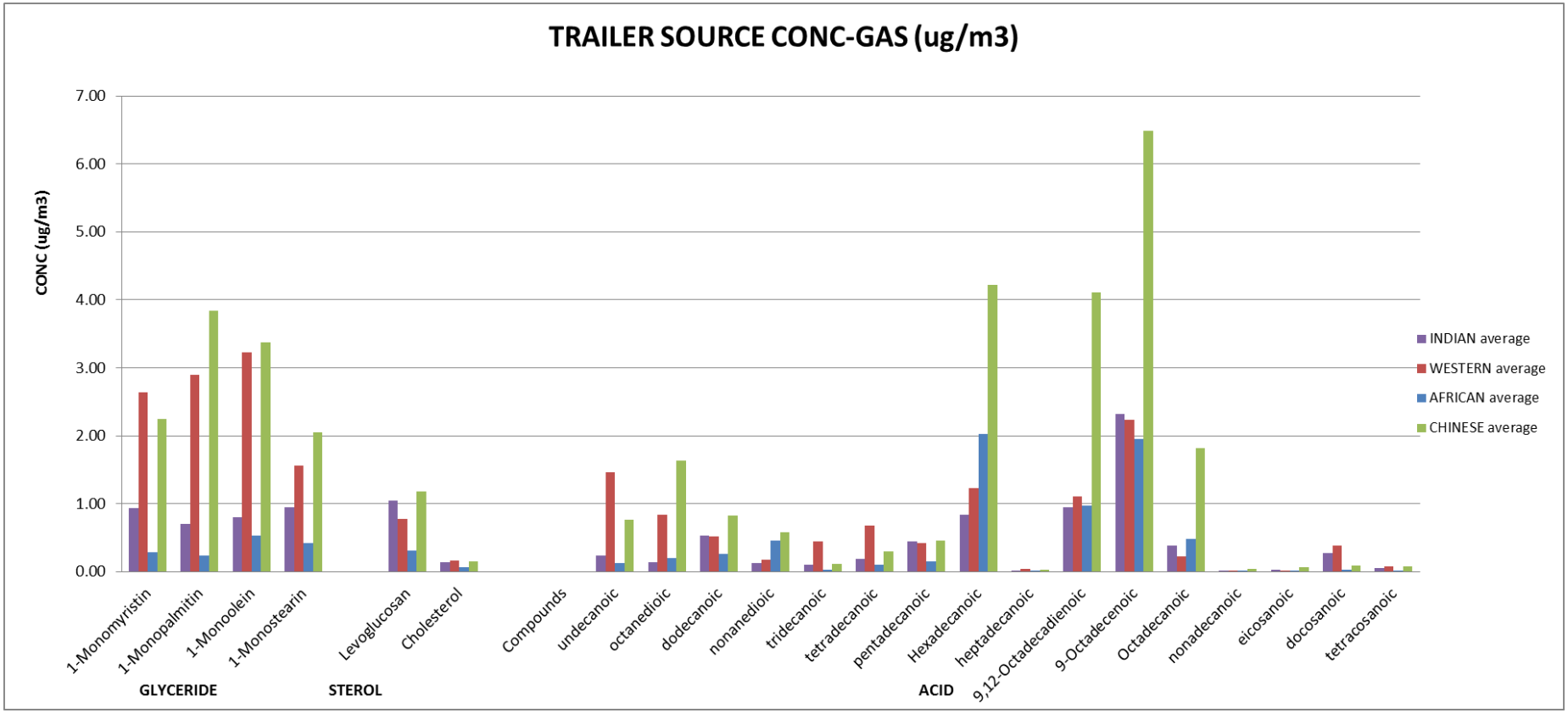


Figure 25 Concentration of compounds (sterol, glyceride and acid) emitted at cooking source (μg/m^3)

Table 23 Concentrations of compounds (Alkane and PAH) emitted at cooking source (µg/m3)

(µg/m3)	INDIAN							WESTERN							AFRICAN							CHINESE						
ALKANES	average	std dev	MIN	25PER	50	75	MAX	average	std dev	MIN	25PER	50	75	MAX	average	std dev	MIN	25PER	50	75	MAX	average	std dev	MIN	25PER	50	75	MAX
Tetracosane	0.67	0.06	0.61	0.63	0.67	0.72	0.74	0.50	0.14	0.36	0.44	0.44	0.50	0.74	0.63	0.34	0.24	0.27	0.86	0.87	0.91	1.28	1.39	0.33	0.42	0.87	1.10	3.70
Pentacosane	0.90	0.24	0.67	0.74	0.86	1.03	1.20	0.62	0.47	0.16	0.41	0.56	0.58	1.41	0.41	0.10	0.31	0.32	0.39	0.49	0.54	0.66	0.23	0.43	0.44	0.68	0.82	0.94
Hexacosane	1.40	0.76	0.45	1.07	1.43	1.77	2.29	1.93	0.93	0.68	1.39	1.94	2.65	2.99	0.49	0.18	0.30	0.32	0.52	0.62	0.71	0.75	0.35	0.51	0.53	0.65	0.69	1.37
Heptacosane	2.71	1.76	1.48	1.82	2.01	2.89	5.32	1.78	0.76	0.69	1.51	1.75	2.26	2.70	0.41	0.07	0.33	0.34	0.42	0.46	0.49	0.87	0.14	0.71	0.81	0.83	0.91	1.08
Octacosane	1.09	0.18	0.93	0.96	1.07	1.21	1.30	1.03	0.36	0.70	0.84	0.95	1.00	1.64	0.38	0.03	0.36	0.37	0.37	0.40	0.42	0.90	0.07	0.81	0.87	0.91	0.92	0.98
Nonacosane	1.22	0.32	0.92	0.97	1.20	1.46	1.57	1.31	0.78	0.83	0.98	1.01	1.03	2.71	0.59	0.08	0.49	0.55	0.57	0.66	0.68	1.33	0.30	1.08	1.20	1.21	1.34	1.84
Triacotane	0.98	0.17	0.82	0.85	0.99	1.12	1.13	0.96	0.27	0.74	0.83	0.88	0.92	1.43	0.35	0.04	0.31	0.32	0.34	0.38	0.40	0.99	0.12	0.88	0.91	0.94	1.04	1.17
Hentriacotane	0.92	0.13	0.80	0.81	0.92	1.03	1.03	0.88	0.17	0.73	0.82	0.82	0.84	1.18	0.34	0.04	0.30	0.31	0.32	0.38	0.39	1.30	0.35	0.82	1.16	1.26	1.53	1.73
Dotriacotane	0.81	0.08	0.73	0.73	0.80	0.88	0.88	0.77	0.14	0.62	0.73	0.75	0.76	1.00	0.30	0.04	0.26	0.28	0.28	0.33	0.34	0.94	0.15	0.79	0.86	0.88	1.02	1.16
Tritriacotane	0.89	0.07	0.82	0.84	0.89	0.95	0.97	1.11	0.74	0.73	0.74	0.81	0.84	2.42	0.40	0.05	0.36	0.36	0.38	0.42	0.48	2.88	0.58	2.27	2.54	2.76	3.03	3.78
Tetratriacotane	0.18	0.01	0.17	0.17	0.17	0.18	0.19	0.16	0.01	0.15	0.15	0.16	0.16	0.17	0.07	0.01	0.06	0.07	0.07	0.08	0.09	0.18	0.02	0.16	0.17	0.18	0.19	0.21
Pentatriacotane	0.64	0.04	0.61	0.61	0.63	0.65	0.69	0.61	0.05	0.56	0.57	0.61	0.62	0.68	0.29	0.05	0.22	0.25	0.32	0.32	0.33	0.90	0.32	0.66	0.68	0.68	1.25	1.25
PAH																												
Benzo[b]fluoranthene	0.54	0.44	0.15	0.16	0.53	0.91	0.96	1.50	0.99	0.13	0.82	1.80	2.27	2.47	0.27	0.18	0.07	0.11	0.32	0.35	0.50	0.90	0.47	0.27	0.77	0.83	1.07	1.58
Benzo[k]fluoranthene	0.32	0.29	0.07	0.07	0.29	0.54	0.63	0.11	0.05	0.07	0.07	0.07	0.16	0.16	0.11	0.04	0.07	0.07	0.11	0.13	0.18	0.35	0.50	0.07	0.07	0.07	0.31	1.22
Benzo[e]pyrene	0.07	0.12	0.01	0.01	0.01	0.07	0.26	0.10	0.18	0.01	0.01	0.02	0.03	0.43	0.14	0.20	0.01	0.01	0.01	0.20	0.48	0.67	0.76	0.01	0.01	0.34	1.48	1.51
Benzo[a]pyrene	0.85	0.76	0.35	0.36	0.55	1.03	1.96	0.97	0.65	0.31	0.54	0.88	1.13	1.99	0.38	0.22	0.15	0.26	0.27	0.58	0.65	1.47	0.96	0.69	1.05	1.22	1.26	3.14
Perylene	0.61	0.74	0.07	0.08	0.35	0.88	1.64	0.76	0.72	0.06	0.13	0.83	0.94	1.84	0.20	0.10	0.03	0.23	0.25	0.26	0.26	1.70	1.27	0.28	1.03	1.10	2.93	3.14
Indeno[123-cd]pyrene	0.68	0.46	0.27	0.29	0.69	1.09	1.09	1.37	0.97	0.24	0.44	1.85	1.92	2.40	0.32	0.15	0.12	0.21	0.35	0.42	0.48	0.97	0.26	0.58	0.92	0.99	1.12	1.26
Dibenz[ah]anthracene	0.96	0.46	0.55	0.58	0.94	1.32	1.42	1.44	0.75	0.49	1.10	1.47	1.60	2.55	0.44	0.17	0.24	0.36	0.37	0.57	0.67	1.96	0.60	1.12	1.56	2.22	2.34	2.56
Picene	0.42	0.42	0.06	0.06	0.38	0.74	0.86	0.68	0.35	0.06	0.75	0.79	0.87	0.94	0.48	0.39	0.06	0.38	0.39	0.46	1.12	0.67	0.19	0.42	0.54	0.69	0.78	0.92
Benzo[ghi]perylene	0.75	0.52	0.30	0.32	0.69	1.12	1.31	1.28	1.21	0.27	0.55	0.89	1.40	3.31	0.23	0.07	0.13	0.19	0.23	0.30	0.31	1.66	0.90	0.49	1.28	1.55	2.08	2.90
Coronene	0.14	0.14	0.02	0.02	0.10	0.22	0.32	1.09	1.12	0.02	0.36	0.65	1.63	2.79	0.33	0.38	0.02	0.18	0.20	0.26	1.00	2.39	1.70	0.14	1.89	2.18	2.89	4.84

Table 24 Concentrations of compounds (glyceride, sterol and acid) emitted at cooking source (µg/m3)

	GAS INDIAN(n=5)							GAS AFR(n=5)							GAS WEST (n=4)							GAS CHINESE (n=4)						
	average	std dev	MIN	25PER	50	75	MAX	average	std dev	MIN	25PER	50	75	MAX	average	std dev	MIN	25PER	50	75	MAX	average	std dev	MIN	25PER	50	75	MAX
GLYCERIDE																												
1-Monomyristin	0.93	0.50	0.39	0.55	0.81	1.45	1.47	0.29	0.17	0.12	0.17	0.25	0.36	0.55	2.64	1.73	1.23	1.33	1.61	4.27	4.76	2.25	1.97	0.57	0.60	2.09	2.62	5.38
1-Monopalmitin	0.70	0.33	0.38	0.48	0.57	0.92	1.16	0.24	0.12	0.11	0.16	0.19	0.37	0.37	2.90	2.74	0.86	1.33	2.17	2.46	7.66	3.84	5.46	0.93	1.30	1.33	2.08	13.58
1-Monoolein	0.80	0.45	0.44	0.51	0.67	0.82	1.55	0.53	0.41	0.19	0.19	0.51	0.53	1.21	3.23	2.81	0.78	0.85	2.07	5.76	6.71	3.37	4.59	1.01	1.23	1.41	1.62	11.58
1-Monostearin	0.95	0.62	0.40	0.52	0.73	1.18	1.93	0.42	0.26	0.16	0.16	0.42	0.59	0.76	1.56	0.26	1.10	1.62	1.66	1.70	1.73	2.05	2.18	0.58	0.58	1.29	2.00	5.82
STEROL																												
Levoglucosan	1.04	0.24	0.72	0.98	0.99	1.12	1.39	0.31	0.14	0.19	0.20	0.23	0.41	0.49	0.78	0.36	0.38	0.56	0.66	0.99	1.29	1.18	0.61	0.57	0.90	1.08	1.17	2.19
Cholesterol	0.14	0.01	0.13	0.14	0.14	0.15	0.16	0.06	0.02	0.05	0.05	0.06	0.08	0.09	0.16	0.05	0.12	0.13	0.15	0.15	0.25	0.15	0.01	0.14	0.15	0.15	0.16	0.17
ACID																												
undecanoic	0.24	0.19	0.11	0.13	0.13	0.27	0.56	0.13	0.14	0.01	0.02	0.07	0.19	0.35	1.47	1.59	0.01	0.13	1.51	2.84	2.84	0.77	1.16	0.16	0.21	0.30	0.32	2.84
octanedioic	0.14	0.11	0.02	0.11	0.12	0.13	0.31	0.20	0.33	0.01	0.02	0.06	0.11	0.78	0.83	0.73	0.12	0.25	0.87	1.46	1.46	1.63	1.92	0.06	0.14	0.67	2.91	4.37
dodecanoic	0.54	0.03	0.51	0.51	0.51	0.57	0.57	0.26	0.36	0.02	0.05	0.13	0.21	0.90	0.52	0.40	0.31	0.31	0.34	0.55	1.12	0.83	1.12	0.14	0.24	0.36	0.57	2.81
nonanedioic	0.13	0.15	0.04	0.05	0.08	0.08	0.39	0.46	0.57	0.02	0.02	0.09	0.99	1.17	0.18	0.13	0.07	0.08	0.15	0.25	0.33	0.58	0.44	0.02	0.20	0.78	0.89	1.00
tridecanoic	0.10	0.06	0.00	0.06	0.14	0.14	0.14	0.02	0.03	0.00	0.00	0.02	0.03	0.07	0.45	0.67	0.01	0.03	0.18	0.60	1.43	0.11	0.20	0.01	0.02	0.03	0.04	0.47
tetradecanoic	0.19	0.12	0.08	0.10	0.14	0.28	0.34	0.10	0.17	0.01	0.01	0.02	0.04	0.40	0.67	1.03	0.07	0.11	0.20	0.76	2.21	0.30	0.29	0.04	0.08	0.16	0.57	0.63
pentadecanoic	0.45	0.17	0.17	0.44	0.52	0.53	0.59	0.15	0.13	0.03	0.08	0.13	0.14	0.38	0.42	0.28	0.21	0.25	0.32	0.49	0.82	0.45	0.31	0.07	0.23	0.51	0.61	0.84
Hexadecanoic	0.84	1.56	0.00	0.15	0.21	0.22	3.63	2.03	2.68	0.01	0.03	0.18	4.68	5.24	1.23	1.27	0.15	0.18	1.05	2.11	2.68	4.22	7.23	0.19	0.20	0.73	3.00	16.98
heptadecanoic	0.01	0.01	0.01	0.01	0.01	0.02	0.02	0.01	0.01	0.00	0.00	0.01	0.02	0.03	0.04	0.05	0.00	0.01	0.02	0.05	0.11	0.03	0.02	0.01	0.02	0.03	0.03	0.07
9,12-Octadecadienoic	0.95	1.22	0.01	0.09	0.11	2.09	2.46	0.97	1.32	0.02	0.04	0.05	1.91	2.82	1.11	1.51	0.10	0.30	0.49	1.30	3.35	4.11	7.88	0.52	0.57	0.59	0.65	18.20
9-Octadecenoic	2.32	2.15	0.36	0.69	1.37	3.91	5.25	1.96	2.67	0.01	0.05	0.11	3.97	5.63	2.24	2.54	0.89	0.94	1.01	2.30	6.04	6.49	8.54	0.69	1.93	2.36	6.14	21.34
Octadecanoic	0.38	0.35	0.00	0.11	0.30	0.75	0.76	0.48	0.27	0.01	0.58	0.58	0.58	0.66	0.23	0.25	0.02	0.02	0.23	0.44	0.44	1.81	2.37	0.02	0.06	0.46	3.15	5.37
nonadecanoic	0.02	0.01	0.01	0.01	0.01	0.02	0.03	0.01	0.01	0.00	0.01	0.01	0.01	0.02	0.02	0.01	0.01	0.02	0.02	0.02	0.03	0.04	0.03	0.02	0.02	0.02	0.07	0.07
eicosanoic	0.02	0.02	0.01	0.01	0.01	0.03	0.04	0.01	0.01	0.00	0.00	0.01	0.02	0.03	0.02	0.01	0.00	0.01	0.01	0.02	0.03	0.06	0.03	0.03	0.04	0.06	0.08	0.11
docosanoic	0.28	0.28	0.04	0.08	0.10	0.53	0.64	0.02	0.02	0.01	0.01	0.02	0.02	0.05	0.38	0.44	0.04	0.18	0.24	0.44	1.03	0.09	0.04	0.05	0.08	0.08	0.11	0.15
tetracosanoic	0.05	0.00	0.04	0.05	0.05	0.05	0.05	0.02	0.01	0.01	0.02	0.02	0.03	0.03	0.07	0.03	0.05	0.05	0.07	0.08	0.11	0.08	0.03	0.06	0.06	0.07	0.07	0.14

Table 25 Indian style cooking concentration (μg/m^3) using gas.

	GAS INDIAN(n=5)						
	average	std dev	MIN	25PER	50	75	MAX
GLYCERIDE							
Compounds							
1-Monomyristin	0.93	0.50	0.39	0.55	0.81	1.45	1.47
1-Monopalmitin	0.70	0.33	0.38	0.48	0.57	0.92	1.16
1-Monoolein	0.80	0.45	0.44	0.51	0.67	0.82	1.55
1-Monostearin	0.95	0.62	0.40	0.52	0.73	1.18	1.93
STEROL							
Levoglucosan	1.04	0.24	0.72	0.98	0.99	1.12	1.39
Cholesterol	0.14	0.01	0.13	0.14	0.14	0.15	0.16
ACID							
undecanoic	0.24	0.19	0.11	0.13	0.13	0.27	0.56
octanedioic	0.14	0.11	0.02	0.11	0.12	0.13	0.31
dodecanoic	0.54	0.03	0.51	0.51	0.51	0.57	0.57
nonanedioic	0.13	0.15	0.04	0.05	0.08	0.08	0.39
tridecanoic	0.10	0.06	0.00	0.06	0.14	0.14	0.14
tetradecanoic	0.19	0.12	0.08	0.10	0.14	0.28	0.34
pentadecanoic	0.45	0.17	0.17	0.44	0.52	0.53	0.59
Hexadecanoic	0.84	1.56	0.00	0.15	0.21	0.22	3.63
heptadecanoic	0.01	0.01	0.01	0.01	0.01	0.02	0.02
9,12-Octadecadienoic	0.95	1.22	0.01	0.09	0.11	2.09	2.46
9-Octadecenoic	2.32	2.15	0.36	0.69	1.37	3.91	5.25
Octadecanoic	0.38	0.35	0.00	0.11	0.30	0.75	0.76
nonadecanoic	0.02	0.01	0.01	0.01	0.01	0.02	0.03
eicosanoic	0.02	0.02	0.01	0.01	0.01	0.03	0.04
docosanoic	0.28	0.28	0.04	0.08	0.10	0.53	0.64
tetracosanoic	0.05	0.00	0.04	0.05	0.05	0.05	0.05
	GAS INDIAN(n=4)						

	average	std dev	MIN	25PER	50	75	MAX
ALKANES							
Tetracosane	0.67	0.06	0.61	0.63	0.67	0.72	0.74
Pentacosane	0.90	0.24	0.67	0.74	0.86	1.03	1.20
Hexacosane	1.40	0.76	0.45	1.07	1.43	1.77	2.29
Heptacosane	2.71	1.76	1.48	1.82	2.01	2.89	5.32
Octacosane	1.09	0.18	0.93	0.96	1.07	1.21	1.30
Nonacosane	1.22	0.32	0.92	0.97	1.20	1.46	1.57
Triacotane	0.98	0.17	0.82	0.85	0.99	1.12	1.13
Hentriacotane	0.92	0.13	0.80	0.81	0.92	1.03	1.03
Dotriacotane	0.81	0.08	0.73	0.73	0.80	0.88	0.88
Tritriacotane	0.89	0.07	0.82	0.84	0.89	0.95	0.97
Tetratriacotane	0.18	0.01	0.17	0.17	0.17	0.18	0.19
Pentatriacotane	0.64	0.04	0.61	0.61	0.63	0.65	0.69
PAH							
Benzo[b]fluoranthene	0.54	0.44	0.15	0.16	0.53	0.91	0.96
Benzo[k]fluoranthene	0.32	0.29	0.07	0.07	0.29	0.54	0.63
Benzo[e]pyrene	0.07	0.12	0.01	0.01	0.01	0.07	0.26
Benzo[a]pyrene	0.85	0.76	0.35	0.36	0.55	1.03	1.96
Perylene	0.61	0.74	0.07	0.08	0.35	0.88	1.64
Indeno[123-cd]pyrene	0.68	0.46	0.27	0.29	0.69	1.09	1.09
Dibenz[ah]anthracene	0.96	0.46	0.55	0.58	0.94	1.32	1.42
Picene	0.42	0.42	0.06	0.06	0.38	0.74	0.86
Benzo[ghi]perylene	0.75	0.52	0.30	0.32	0.69	1.12	1.31
Coronene	0.14	0.14	0.02	0.02	0.10	0.22	0.32

Table 26 Chinese style cooking concentration (μg/m^3) using gas.

	GAS CHINESE (n=4)						
	average	std dev	MIN	25PER	50	75	MAX
GLYCERIDE							
1-Monomyristin	2.25	1.97	0.57	0.60	2.09	2.62	5.38
1-Monopalmitin	3.84	5.46	0.93	1.30	1.33	2.08	13.58
1-Monoolein	3.37	4.59	1.01	1.23	1.41	1.62	11.58
1-Monostearin	2.05	2.18	0.58	0.58	1.29	2.00	5.82
STEROL							
Levoglucosan	1.18	0.61	0.57	0.90	1.08	1.17	2.19
Cholesterol	0.15	0.01	0.14	0.15	0.15	0.16	0.17
ACID							
undecanoic	0.77	1.16	0.16	0.21	0.30	0.32	2.84
octanedioic	1.63	1.92	0.06	0.14	0.67	2.91	4.37
dodecanoic	0.83	1.12	0.14	0.24	0.36	0.57	2.81
nonanedioic	0.58	0.44	0.02	0.20	0.78	0.89	1.00
tridecanoic	0.11	0.20	0.01	0.02	0.03	0.04	0.47
tetradecanoic	0.30	0.29	0.04	0.08	0.16	0.57	0.63
pentadecanoic	0.45	0.31	0.07	0.23	0.51	0.61	0.84
Hexadecanoic	4.22	7.23	0.19	0.20	0.73	3.00	16.98
heptadecanoic	0.03	0.02	0.01	0.02	0.03	0.03	0.07
9,12-Octadecadienoic	4.11	7.88	0.52	0.57	0.59	0.65	18.20
9-Octadecenoic	6.49	8.54	0.69	1.93	2.36	6.14	21.34
Octadecanoic	1.81	2.37	0.02	0.06	0.46	3.15	5.37
nonadecanoic	0.04	0.03	0.02	0.02	0.02	0.07	0.07
eicosanoic	0.06	0.03	0.03	0.04	0.06	0.08	0.11
docosanoic	0.09	0.04	0.05	0.08	0.08	0.11	0.15
tetracosanoic	0.08	0.03	0.06	0.06	0.07	0.07	0.14
	GAS CHINESE (n=5)						
	average	std dev	MIN	25PER	50	75	MAX
ALKANES							
Tetracosane	1.28	1.39	0.33	0.42	0.87	1.10	3.70
Pentacosane	0.66	0.23	0.43	0.44	0.68	0.82	0.94

Hexacosane	0.75	0.35	0.51	0.53	0.65	0.69	1.37
Heptacosane	0.87	0.14	0.71	0.81	0.83	0.91	1.08
Octacosane	0.90	0.07	0.81	0.87	0.91	0.92	0.98
Nonacosane	1.33	0.30	1.08	1.20	1.21	1.34	1.84
Triacotane	0.99	0.12	0.88	0.91	0.94	1.04	1.17
Hentriacotane	1.30	0.35	0.82	1.16	1.26	1.53	1.73
Dotriacotane	0.94	0.15	0.79	0.86	0.88	1.02	1.16
Tritriacotane	2.88	0.58	2.27	2.54	2.76	3.03	3.78
Tetratriacotane	0.18	0.02	0.16	0.17	0.18	0.19	0.21
Pentatriacotane	0.90	0.32	0.66	0.68	0.68	1.25	1.25
PAH							
Benzo[b]fluoranthene	0.90	0.47	0.27	0.77	0.83	1.07	1.58
Benzo[k]fluoranthene	0.35	0.50	0.07	0.07	0.07	0.31	1.22
Benzo[e]pyrene	0.67	0.76	0.01	0.01	0.34	1.48	1.51
Benzo[a]pyrene	1.47	0.96	0.69	1.05	1.22	1.26	3.14
Perylene	1.70	1.27	0.28	1.03	1.10	2.93	3.14
Indeno[123-cd]pyrene	0.97	0.26	0.58	0.92	0.99	1.12	1.26
Dibenz[ah]anthracene	1.96	0.60	1.12	1.56	2.22	2.34	2.56
Picene	0.67	0.19	0.42	0.54	0.69	0.78	0.92
Benzo[ghi]perylene	1.66	0.90	0.49	1.28	1.55	2.08	2.90
Coronene	1.47	0.96	0.69	1.05	1.22	1.26	3.14

Table 27 African style cooking concentration (µg/m^3) using gas

	GAS AFR(n=5)						
	average	std dev	MIN	25PER	50	75	MAX
GLYCERIDE							
1-Monomyristin	0.29	0.17	0.12	0.17	0.25	0.36	0.55
1-Monopalmitin	0.24	0.12	0.11	0.16	0.19	0.37	0.37
1-Monoolein	0.53	0.41	0.19	0.19	0.51	0.53	1.21
1-Monostearin	0.42	0.26	0.16	0.16	0.42	0.59	0.76
STEROL							
Levoglucosan	0.31	0.14	0.19	0.20	0.23	0.41	0.49
Cholesterol	0.06	0.02	0.05	0.05	0.06	0.08	0.09
ACID							
undecanoic	0.13	0.14	0.01	0.02	0.07	0.19	0.35
octanedioic	0.20	0.33	0.01	0.02	0.06	0.11	0.78
dodecanoic	0.26	0.36	0.02	0.05	0.13	0.21	0.90
nonanedioic	0.46	0.57	0.02	0.02	0.09	0.99	1.17
tridecanoic	0.02	0.03	0.00	0.00	0.02	0.03	0.07
tetradecanoic	0.10	0.17	0.01	0.01	0.02	0.04	0.40
pentadecanoic	0.15	0.13	0.03	0.08	0.13	0.14	0.38
Hexadecanoic	2.03	2.68	0.01	0.03	0.18	4.68	5.24
heptadecanoic	0.01	0.01	0.00	0.00	0.01	0.02	0.03
9,12-Octadecadienoic	0.97	1.32	0.02	0.04	0.05	1.91	2.82
9-Octadecenoic	1.96	2.67	0.01	0.05	0.11	3.97	5.63
Octadecanoic	0.48	0.27	0.01	0.58	0.58	0.58	0.66
nonadecanoic	0.01	0.01	0.00	0.01	0.01	0.01	0.02
eicosanoic	0.01	0.01	0.00	0.00	0.01	0.02	0.03
docosanoic	0.02	0.02	0.01	0.01	0.02	0.02	0.05
tetracosanoic	0.02	0.01	0.01	0.02	0.02	0.03	0.03
	GAS AFR(n=5)						
	average	std dev	MIN	25PER	50	75	MAX
ALKANES							
Tetracosane	0.63	0.34	0.24	0.27	0.86	0.87	0.91
Pentacosane	0.41	0.10	0.31	0.32	0.39	0.49	0.54
Hexacosane	0.49	0.18	0.30	0.32	0.52	0.62	0.71

Heptacosane	0.41	0.07	0.33	0.34	0.42	0.46	0.49
Octacosane	0.38	0.03	0.36	0.37	0.37	0.40	0.42
Nonacosane	0.59	0.08	0.49	0.55	0.57	0.66	0.68
Triacotane	0.35	0.04	0.31	0.32	0.34	0.38	0.40
Hentriacotane	0.34	0.04	0.30	0.31	0.32	0.38	0.39
Dotriacotane	0.30	0.04	0.26	0.28	0.28	0.33	0.34
Tritriacotane	0.40	0.05	0.36	0.36	0.38	0.42	0.48
Tetratriacotane	0.07	0.01	0.06	0.07	0.07	0.08	0.09
Pentatriacotane	0.29	0.05	0.22	0.25	0.32	0.32	0.33
PAH							
Benzo[b]fluoranthene	0.27	0.18	0.07	0.11	0.32	0.35	0.50
Benzo[k]fluoranthene	0.11	0.04	0.07	0.07	0.11	0.13	0.18
Benzo[e]pyrene	0.14	0.20	0.01	0.01	0.01	0.20	0.48
Benzo[a]pyrene	0.38	0.22	0.15	0.26	0.27	0.58	0.65
Perylene	0.20	0.10	0.03	0.23	0.25	0.26	0.26
Indeno[123-cd]pyrene	0.32	0.15	0.12	0.21	0.35	0.42	0.48
Dibenz[ah]anthracene	0.44	0.17	0.24	0.36	0.37	0.57	0.67
Picene	0.48	0.39	0.06	0.38	0.39	0.46	1.12
Benzo[ghi]perylene	0.23	0.07	0.13	0.19	0.23	0.30	0.31
Coronene	0.33	0.38	0.02	0.18	0.20	0.26	1.00

Table 28 Western style cooking concentration(μg/m^3) using gas

	GAS WEST (n=4)						
	average	std dev	MIN	25PER	50	75	MAX
GLYCERIDE							
1-Monomyristin	2.64	1.73	1.23	1.33	1.61	4.27	4.76
1-Monopalmitin	2.90	2.74	0.86	1.33	2.17	2.46	7.66
1-Monoolein	3.23	2.81	0.78	0.85	2.07	5.76	6.71
1-Monostearin	1.56	0.26	1.10	1.62	1.66	1.70	1.73
STEROL							
Levoglucosan	0.78	0.36	0.38	0.56	0.66	0.99	1.29
Cholesterol	0.16	0.05	0.12	0.13	0.15	0.15	0.25
ACID							
undecanoic	1.47	1.59	0.01	0.13	1.51	2.84	2.84
octanedioic	0.83	0.73	0.12	0.25	0.87	1.46	1.46
dodecanoic	0.52	0.40	0.31	0.31	0.34	0.55	1.12
nonanedioic	0.18	0.13	0.07	0.08	0.15	0.25	0.33
tridecanoic	0.45	0.67	0.01	0.03	0.18	0.60	1.43
tetradecanoic	0.67	1.03	0.07	0.11	0.20	0.76	2.21
pentadecanoic	0.42	0.28	0.21	0.25	0.32	0.49	0.82
Hexadecanoic	1.23	1.27	0.15	0.18	1.05	2.11	2.68
heptadecanoic	0.04	0.05	0.00	0.01	0.02	0.05	0.11
9,12-Octadecadienoic	1.11	1.51	0.10	0.30	0.49	1.30	3.35
9-Octadecenoic	2.24	2.54	0.89	0.94	1.01	2.30	6.04
Octadecanoic	0.23	0.25	0.02	0.02	0.23	0.44	0.44
nonadecanoic	0.02	0.01	0.01	0.02	0.02	0.02	0.03
eicosanoic	0.02	0.01	0.00	0.01	0.01	0.02	0.03
docosanoic	0.38	0.44	0.04	0.18	0.24	0.44	1.03
tetracosanoic	0.07	0.03	0.05	0.05	0.07	0.08	0.11
	GAS WEST (n=5)						
	average	std dev	MIN	25PER	50	75	MAX
ALKANES							
Tetracosane	0.50	0.14	0.36	0.44	0.44	0.50	0.74
Pentacosane	0.62	0.47	0.16	0.41	0.56	0.58	1.41

Hexacosane	1.93	0.93	0.68	1.39	1.94	2.65	2.99
Heptacosane	1.78	0.76	0.69	1.51	1.75	2.26	2.70
Octacosane	1.03	0.36	0.70	0.84	0.95	1.00	1.64
Nonacosane	1.31	0.78	0.83	0.98	1.01	1.03	2.71
Triacotane	0.96	0.27	0.74	0.83	0.88	0.92	1.43
Hentriacotane	0.88	0.17	0.73	0.82	0.82	0.84	1.18
Dotriacotane	0.77	0.14	0.62	0.73	0.75	0.76	1.00
Tritriacotane	1.11	0.74	0.73	0.74	0.81	0.84	2.42
Tetratriacotane	0.16	0.01	0.15	0.15	0.16	0.16	0.17
Pentatriacotane	0.61	0.05	0.56	0.57	0.61	0.62	0.68
PAH							
Benzo[b]fluoranthene	1.50	0.99	0.13	0.82	1.80	2.27	2.47
Benzo[k]fluoranthene	0.11	0.05	0.07	0.07	0.07	0.16	0.16
Benzo[e]pyrene	0.10	0.18	0.01	0.01	0.02	0.03	0.43
Benzo[a]pyrene	0.97	0.65	0.31	0.54	0.88	1.13	1.99
Perylene	0.76	0.72	0.06	0.13	0.83	0.94	1.84
Indeno[123-cd]pyrene	1.37	0.97	0.24	0.44	1.85	1.92	2.40
Dibenz[ah]anthracene	1.44	0.75	0.49	1.10	1.47	1.60	2.55
Picene	0.68	0.35	0.06	0.75	0.79	0.87	0.94
Benzo[ghi]perylene	1.28	1.21	0.27	0.55	0.89	1.40	3.31
Coronene	1.09	1.12	0.02	0.36	0.65	1.63	2.79

Table 29 Average concentrations of compounds emitted at source Indian cooking styles using electric (µg/m^3).

	ELEC-TC IND(n=4)						
	average	std dev	MIN	25PER	50	75	MAX
GLYCERIDE							
1-Monomyristin	0.43	0.06	0.35	0.42	0.45	0.46	0.48
1-Monopalmitin	0.51	0.12	0.36	0.47	0.51	0.55	0.66
1-Monoolein	0.46	0.06	0.40	0.40	0.45	0.51	0.52
1-Monostearin	0.52	0.16	0.32	0.45	0.54	0.62	0.69
STEROL							
Levoglucosan	0.19	0.04	0.15	0.16	0.18	0.21	0.24
Cholesterol	0.14	0.02	0.12	0.14	0.14	0.15	0.17
ACID							
undecanoic	0.35	0.25	0.13	0.13	0.35	0.56	0.56
octanedioic	0.13	0.01	0.12	0.12	0.13	0.13	0.13
dodecanoic	0.41	0.24	0.05	0.40	0.51	0.53	0.57
nonanedioic	0.05	0.01	0.04	0.04	0.05	0.05	0.05
tridecanoic	0.07	0.06	0.00	0.05	0.06	0.08	0.14
tetradecanoic	0.15	0.09	0.10	0.10	0.10	0.15	0.28
pentadecanoic	0.31	0.26	0.03	0.13	0.30	0.48	0.59
Hexadecanoic	0.35	0.16	0.22	0.23	0.31	0.42	0.55
heptadecanoic	0.01	0.01	0.01	0.01	0.01	0.02	0.02
9,12-Octadecadienoic	0.25	0.15	0.09	0.17	0.22	0.30	0.45
9-Octadecenoic	0.29	0.27	0.02	0.18	0.23	0.34	0.67
Octadecanoic	1.13	1.23	0.11	0.59	0.75	1.30	2.91
nonadecanoic	0.03	0.00	0.03	0.03	0.03	0.03	0.03
eicosanoic	0.01	0.00	0.01	0.01	0.01	0.01	0.01
docosanoic	0.02	0.00	0.02	0.02	0.02	0.02	0.03
tetracosanoic	0.04	0.00	0.03	0.03	0.03	0.04	0.04

Table 30 Average concentrations of compounds emitted at source African cooking styles using electric (µg/m^3).

	ELEC AFR (n=4)						
	average	std dev	MIN	25PER	50	75	MAX
GLYCERIDE							
1-Monomyristin	0.22	0.04	0.18	0.20	0.21	0.23	0.27
1-Monopalmitin	0.22	0.01	0.20	0.21	0.22	0.23	0.23
1-Monoolein	0.24	0.05	0.19	0.21	0.23	0.26	0.31
1-Monostearin	0.22	0.06	0.17	0.18	0.21	0.25	0.30
STEROL							
Levoglucosan	0.11	0.02	0.08	0.09	0.11	0.12	0.13
Cholesterol	0.08	0.02	0.06	0.07	0.08	0.10	0.11
ACID							
undecanoic	0.08	0.00	0.08	0.08	0.08	0.08	0.08
octanedioic	0.12	0.01	0.11	0.11	0.12	0.13	0.13
dodecanoic	0.07	0.04	0.05	0.05	0.05	0.07	0.13
nonanedioic	0.28	0.48	0.02	0.02	0.06	0.32	0.99
tridecanoic	0.03	0.03	0.00	0.01	0.02	0.04	0.07
tetradecanoic	0.03	0.02	0.01	0.01	0.03	0.04	0.06
pentadecanoic	0.08	0.07	0.00	0.06	0.08	0.10	0.17
Hexadecanoic	0.29	0.10	0.15	0.26	0.30	0.32	0.40
heptadecanoic	0.04	0.03	0.02	0.02	0.04	0.07	0.07
9,12-Octadecadienoic	7.83	15.19	0.16	0.20	0.28	7.91	30.61
9-Octadecenoic	2.45	4.32	0.06	0.22	0.40	2.63	8.92
Octadecanoic	0.74	1.45	0.02	0.02	0.02	0.74	2.91
nonadecanoic	0.03	0.01	0.02	0.02	0.03	0.03	0.03
eicosanoic	0.42	0.27	0.01	0.42	0.55	0.55	0.55
docosanoic	0.02	0.01	0.01	0.01	0.01	0.02	0.03
tetracosanoic	0.03	0.03	0.02	0.02	0.02	0.03	0.08

Table 31 Average concentrations of compounds emitted at source using Western cooking styles using electric (µg/m^3).

	ELEC WEST (n=4)						
	average	std dev	MIN	25PER	50	75	MAX
GLYCERIDE							
1-Monomyristin	0.69	0.30	0.35	0.51	0.69	0.87	1.03
1-Monopalmitin	0.73	0.39	0.36	0.43	0.69	0.99	1.20
1-Monoolein	0.69	0.46	0.35	0.46	0.52	0.75	1.37
1-Monostearin	0.64	0.52	0.29	0.39	0.43	0.68	1.41
STEROL							
Levoglucosan	0.37	0.44	0.11	0.12	0.17	0.41	1.03
Cholesterol	0.19	0.09	0.11	0.12	0.16	0.23	0.31
ACID							
undecanoic	0.38	0.05	0.35	0.35	0.36	0.39	0.45
octanedioic	0.07	0.00	0.06	0.06	0.07	0.07	0.07
dodecanoic	0.47	0.49	0.05	0.05	0.47	0.90	0.90
nonanedioic	0.30	0.58	0.02	0.02	0.02	0.30	1.17
tridecanoic	0.03	0.03	0.00	0.01	0.02	0.04	0.07
tetradecanoic	0.11	0.19	0.01	0.02	0.02	0.12	0.40
pentadecanoic	0.14	0.17	0.02	0.02	0.07	0.19	0.38
Hexadecanoic	0.17	0.10	0.03	0.14	0.21	0.24	0.24
heptadecanoic	0.11	0.08	0.02	0.07	0.09	0.12	0.22
9,12-Octadecadienoic	0.18	0.16	0.10	0.10	0.10	0.18	0.41
9-Octadecenoic	1.22	1.86	0.09	0.12	0.41	1.51	3.97
Octadecanoic	0.62	0.05	0.58	0.58	0.62	0.66	0.66
nonadecanoic	0.01	0.00	0.01	0.01	0.01	0.01	0.01
eicosanoic	0.02	0.00	0.02	0.02	0.02	0.02	0.02
docosanoic	0.02	0.01	0.02	0.02	0.02	0.02	0.03
tetracosanoic	0.04	0.01	0.03	0.04	0.04	0.05	0.05

Table 32 Average concentrations of compounds emitted at source Chinese cooking styles using electric (µg/m^3).

	ELECT CHIN (n=4 sterol)) (n=3 acids)						
	average	std dev	MIN	25PER	50	75	MAX
GLYCERIDE							
1-Monomyristin	0.63	0.22	0.44	0.52	0.57	0.68	0.95
1-Monopalmitin	0.80	0.37	0.36	0.64	0.79	0.95	1.26
1-Monoolein	0.88	0.48	0.46	0.55	0.76	1.08	1.53
1-Monostearin	0.76	0.35	0.40	0.53	0.71	0.94	1.21
STEROL							
Levoglucosan	0.42	0.20	0.28	0.28	0.35	0.49	0.69
Cholesterol	0.21	0.07	0.16	0.17	0.18	0.21	0.31
ACID							
undecanoic	2.97	0.23	2.84	2.84	2.84	3.04	3.24
octanedioic							
dodecanoic							
nonanedioic							
tridecanoic							
tetradecanoic							
pentadecanoic	0.03	0.01	0.03	0.03	0.03	0.03	0.04
Hexadecanoic	0.89	0.44	0.47	0.66	0.85	1.10	1.35
heptadecanoic							
9,12-Octadecadienoic	1.94	0.85	0.97	1.63	2.29	2.42	2.55
9-Octadecenoic	1.45	1.15	0.12	1.12	2.11	2.11	2.11
Octadecanoic	2.00	2.93	0.16	0.31	0.46	2.92	5.37
nonadecanoic							
eicosanoic	0.03	0.02	0.01	0.02	0.03	0.04	0.04
docosanoic	0.03	0.01	0.03	0.03	0.04	0.04	0.04
tetracosanoic	0.06	0.01	0.05	0.06	0.06	0.06	0.06

An analysis of the various source profiles against themselves to observe how they relate with each other is presented below in Table 33. The reason for this analysis is to determine if the various cooking styles profiles obtained differ greatly from one another, this knowledge would be useful to know if a single profile can represent all cooking styles considered. This could be beneficial knowledge for source input in modelling and this would provide more insight to the accuracy of general conclusions drawn from selecting a single profile to represent all possible cooking methods.

The Spearman's rank correlation analysis for the various groups of compounds against the different types of cooking is analysed; Western and Indian cooking are observed to have a strong correlation for alkanes with r_s values of 0.902. Chinese cooking and Indian cooking are the least correlated or similar in terms of alkane with r_s of 0.021.

For PAH concentration it is observed that the Indian and western style of cooking are the most correlated even though the degree of correlation is much less than in the case of alkanes (0.61). The Chinese cooking PAH concentrations are found to have an r_s value of 0.5 when compared with Indian cooking similar to the correlation of Chinese and African cooking.

The correlation analysis of acids for all cooking styles yields very good r_s values for all the profiles, ranging between 0.8 (Western and Indian) and 0.9 (Indian and Chinese).

When the correlations of all compounds groups are considered together, the most similar profiles are the Western and Indian profiles (with r_s values of 0.9, 0.6 and 0.8 for Alkanes, PAH and acid respectively). Chinese and Indian profiles have weak correlation in terms of alkane and PAH compounds but have high correlation in terms of the concentration of acids (0.02, 0.5 and 0.9 respectively) as such will be considered having the weakest correlation considering all compounds.

African cooking have low correlation with Chinese and Indian profiles for alkanes (0.21 and 0.5) and for PAH the r_s values are higher but still correlation level is quite low (0.51 and 0.5 respectively).

Western cooking appears to be the profile that correlates the best with all the profiles with only a weak correlation with Chinese alkane concentration (0.2). All other values fall within the range 0.5-0.8 considering all group of compounds (alkanes, PAH and acids): example r_s values of 0.77 Western and Indian acid,0.71 western and African acid, 0.81 western and Chinese acid, PAH – 0.61 Western And Indian, 0.52 Western and African, 0.62 Western and Chinese

Table 33 Correlation of various groups of compounds among the different cooking styles.

ALKANE

Correlations

			INDIAN	WESTERN	AFRICAN	CHINESE
Spearman's rho	INDIAN	Correlation Coefficient	1.000	.902**	.510	.021
		Sig. (2-tailed)	.	.000	.090	.948
		N	12	12	12	12
	WESTERN	Correlation Coefficient	.902**	1.000	.441	.189
		Sig. (2-tailed)	.000	.	.152	.557
		N	12	12	12	12
	AFRICAN	Correlation Coefficient	.510	.441	1.000	.210
		Sig. (2-tailed)	.090	.152	.	.513
		N	12	12	12	12
	CHINESE	Correlation Coefficient	.021	.189	.210	1.000
		Sig. (2-tailed)	.948	.557	.513	.
		N	12	12	12	12

**. Correlation is significant at the 0.01 level (2-tailed).

PAH

Correlations

			INDIAN	WESTERN	AFRICAN	CHINESE
Spearman's rho	INDIAN	Correlation Coefficient	1.000	.608*	.483	.490
		Sig. (2-tailed)	.	.036	.112	.106
		N	12	12	12	12
	WESTERN	Correlation Coefficient	.608*	1.000	.524	.623*
		Sig. (2-tailed)	.036	.	.080	.030
		N	12	12	12	12
	AFRICAN	Correlation Coefficient	.483	.524	1.000	.508
		Sig. (2-tailed)	.112	.080	.	.092
		N	12	12	12	12
	CHINESE	Correlation Coefficient	.490	.623*	.508	1.000
		Sig. (2-tailed)	.106	.030	.092	.
		N	12	12	12	12

*. Correlation is significant at the 0.05 level (2-tailed).

ACID

Correlations

			INDIAN	WESTERN	AFRICAN	CHINESE
Spearman's rho	INDIAN	Correlation Coefficient	1.000	.770**	.856**	.870**
		Sig. (2-tailed)	.	.000	.000	.000
		N	16	16	16	16
	WESTERN	Correlation Coefficient	.770**	1.000	.708**	.811**
		Sig. (2-tailed)	.000	.	.002	.000
		N	16	16	16	16
	AFRICAN	Correlation Coefficient	.856**	.708**	1.000	.965**
		Sig. (2-tailed)	.000	.002	.	.000
		N	16	16	16	16
	CHINESE	Correlation Coefficient	.870**	.811**	.965**	1.000
		Sig. (2-tailed)	.000	.000	.000	.
		N	16	16	16	16

**. Correlation is significant at the 0.01 level (2-tailed).

ANOVA was carried out for the concentrations across the group using the four types of cooking. It was found that the means were significantly different with a sig (0.001)

3.5 Diagnostic ratio and cooking.

Diagnostic ratio is a binary ratio method for source identification which involves comparing ratios of pairs of frequently found compounds emitted to distinguish between different known sources. It is usually used with caution as it is often difficult to discriminate between some sources and also its interpretation depends on the ratio considered and profile chosen so it can vary in presence of highly reactive compounds and thus can introduce bias.

Some studies have shown similar diagnostic ratio for different sources for example PAH ratio between 0.4 and 0.5 for FLU/(FLU + PYR) may indicate possible sources such as cement production, fertiliser production, diesel combustion, metal manufacturing, and road dust, while another diagnostic ratio from the same data set may show a strong variation for a particular source, BbF/BkF = 2.5–2.9 for aluminium smelter emissions (Alam et al., 2013; Manoli et al, 2004). It has also been identified that variation in atmospheric processes and combustion conditions affect the emission and degradation of individual compounds as such can affect ratios obtained by these compounds (Katsoyiannis et al. 2011). This makes these ratios reliability questionable however this is minimised by selection of compounds with similar physicochemical properties. Atmospheric processes can hinder these diagnostic ratios as individual PAH compounds have

different atmospheric lifetimes and reactivities (Atkinson and Arey, 2007; Arey, 1998; (Alam et al., 2013).

Most diagnostic ratios involve pairs of compounds with the same molar mass and similar physicochemical properties so should undergo similar environmental fate processes. The PAH emission profile for a given source depends on the processes producing the PAHs (Manoli et al., 2004). During processes like wood burning which involve low temperature, low molecular weight PAHs are usually formed while high temperature combustion eg. burning of fuels in engines, result in the emission of higher molecular weight PAH compounds (Mostert et al., 2010).

At high temperatures organic compounds are cracked to reactive radicals, which react to form stable PAHs during pyrosynthesis. The formed PAHs are less alkylated and their molecules and contain more aromatic rings than petrogenic PAHs (Hwang et al., 2003). PAH diagnostic ratios have been used to distinguish diesel and gasoline combustion emission (Ravindra et al., 2008), different crude oil processing products and biomass burning processes, including bush, savanna and grass fires (Yunker et al., 2002; Galarneau, 2008; Tobiszewski and Namieśnik, 2012). Table 34 A. and B presents the diagnostic ratio of various PAHs for different combustion sources from some previous studies and Table 34 C from this study.

Plots of concentrations of different markers against each other to see how well correlated they are as shown in Figure 26 , has been a useful tool to determine diagnostic ratios. Robinson et al., (2006) made plots of various ambient species, they focused on only five important markers for cooking; n-hexadecanoic (palmitic) acid, n-octadecanoic (stearic) acid, 9-hexadecenoic (palmitoleic) acid, 9-octadecenoic (oleic) acid, and cholesterol. Oleic and palmitoleic acid concentrations as well as stearic and palmitic acid concentrations which were well correlated with a slope of one implying a single dominant source for the alkenoic acids only a slight correlation was observed between cholesterol and palmitic acid but the scatter was comparable to measurement uncertainty so the sources could have been the same. Saturated and unsaturated acid however when plotted against

themselves showed no correlation (palmitic acid against palmitoleic acid) (Robinson et al., 2006). It was concluded that these acids had different dominant sources it was assumed they were chemically stable. Ratio –ratio plots aid in the inference of potential source profiles, these plots should be examined using different ratio species and different combinations of source specific markers to develop a good understanding of ambient data and source profiles. Robinson et al., 2006 made some of such plots of acids (two alkanoinc and alkenoic) normalised by cholesterol and observed a good correlations in ambient data by displaying well organised ratio to ratio plots. Normalisation is a general approach to reduce the anomalies in large data sets. Scatters along the diagonals of the plots can be attributed to measurement uncertainty or the variability of emissions of species. The reference specie used to normalise the concentrations of the two target species affects the exact organisation of data in a ratio-ratio plot and by changing this reference specie does not alter the likely conclusion about the source profile combination (Robinson et al., 2006). The change however causes the location of both the source profiles and ambient data in the plots to shift. Cholesterol has been found to be a good reference for food cooking markers and so was generally used.

Diagnostic ratios for PAHs, such as BeP/(BeP + BaP), IND/(IND + BghiP), Cor/BeP and BghiP/BeP can be useful in the investigation of their origins and so aid in the identification of the possible emission sources in air samples. See et al., (2006) used this technique in combination with other statistical methods and it is discussed. The ratios were also calculated to provide insight on the origins as well as markers or tracers of pollution source. The ratios of Phe/(ant+Phe) (structural isomers of molecular weight MW=178), Flt/(Flt+Pyr)(MW=202), BaA/(BaA+Chr)(MW=228) and Ind/(Ind+BPe)(MW=276) were evaluated based on mean concentrations (See et al 2008). These ratios are useful to compare, determine and confirm the PAHs measured are from cooking sources as other common sources of PAH exist. Table 34 shows some diagnostic ratios obtained for culinary techniques and from vehicle emissions. Miguel and Pereira in 1989 found that generally PAHs from

petroleum source had ratios of FLU/(FLU+PYR) < 0.2, while 0.4–0.5 for combustion of fuel (tail gas from vehicles) and > 0.5 for grass, wood and coal combustion Sheesley et al.,(2003) found that FLU/(FLU+PYR) ratios for PAHs from the burning of rice straw in Asia was 0.51 while in a Chinese tunnel mean ratio of FLU/(FLU+PYR) was about 0.4(Sheesley et al., 2003). The FLU/(FLU+PYR) ratio of 0.53 was obtained in China and was attributed to the burning of bituminous coal by (Liu et al., 2009), it was also found that the FLU/(FLU+PYR) ratios of 0.40 to 0.58 was obtained in the study by Gu et al., (2010) with PAHs in $PM_{2.5}$ obtained in urban Shanghai being observed to be from mixed sources of coal/biomass burning and vehicle emissions(Gu et al., 2010). In Table 34 (C) the diagnostic ratio, from this study,of various PAHs for the four cooking styles IND(IND+BghiP are very similar for all the cooking styles. BghiP/BeP is high for Indian and Western style cooking and lower in the African and Chinese cooking profile.

Comparism of diagnostic ratios of Table 34 A and B (ratios obtained for culinary techniques and from vehicle emissions from literature) show that ratios from See et al. (2006) and See and Balasubramanian (2008) are relatively unaffected by type of cooking and there is some overlap in cooking ratios with those from traffic, making quantitative differentiation impossible.

Some size resolved source apportionment studies have used molecular marker to organic carbon ratios for chemical signature for source contribution identifications and good tracers have been identified for molecular markers with similar size distribution for EC and OC (Kleeman et al. 2008).The calculation of correlation coefficient (R^2) of concentrations of two species can be analysed for similarity of the size distributions. Kleeman et al., 2008 found that the size distribution for cholesterol was highly correlated (R2>0.9) with both OC and EC size distribution further confirming that cholesterol can serve as an appropriate tracer for meat cooking contributions. The most abundant PAH measured in emission from meat cooking was phenanthrene with small concentrations of fluoranthene and pyrene, however higher ambient concentrations of these were observed from other sources such as diesel engines (Kleeman et al., 2008).

The diagnostic ratios plotted against each other in Figure 27 shows that across the cooking profiles there exists a clearer difference for ratios of Corene/BeP and BghiP/BeP. BeP/BeP+BaP) and are very similar for all the cooking styles IND(IND+BghiP).

Table 35 shows the diagnostic ratio for selected acids for this study. It also shows these ratios obtained from literature. A similar trend is observed across all the ratio of acids however higher values are obtained for oleic to steric acid ratio in this study as compares to previous studies a ratio of 9.5 is obtained for western style cooking.

When analysing the diagnostic ratio for acid across various studies including data from this study, in Table 35 Figure 27 and Figure 28, oleic/linoleic acids ratio as well as oleic/stearic acid ratios were observed to differ across the concentrations analysed with good correlation for cholesteral/ palmitic acid and stearic /palmitic acid. Oleic/linoleic ratio was highest for Indian cooking (2.43) and lowest for Chinese cooking (1.58). oleic/stearic acid ratios were generally higher compared to other diagnostic ratios with western cooking having the highest value of 9.75 followed by Indian cooking and 4.05 bring the ratio for African cooking. The lowest ratio for oleic to stearic acid was observed for Chinese style cooking with a value of 3.58.

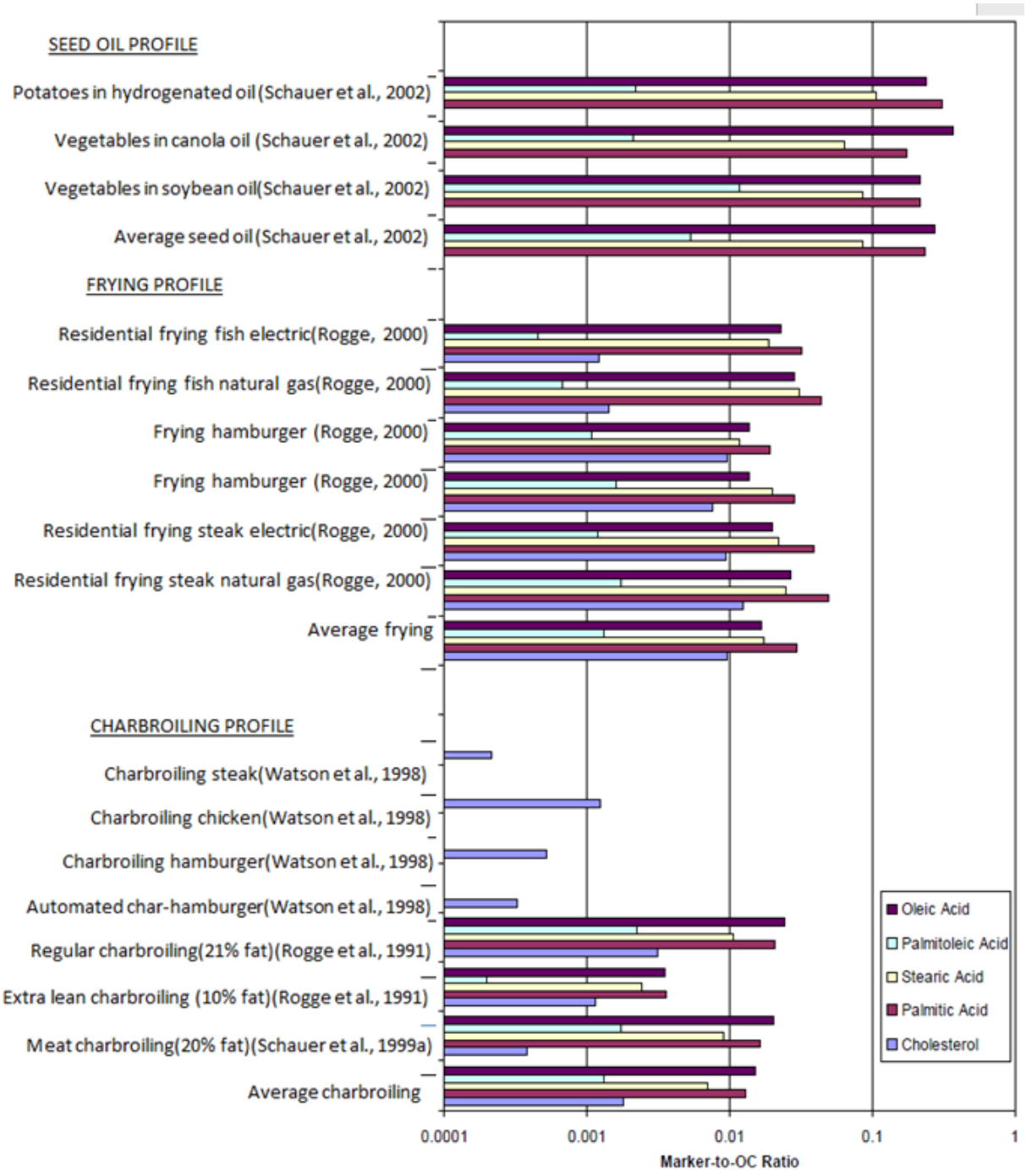


Figure 26 Marker to OC ratio for meat cooking profiles (Robinson et al., 2006)

Table 34 Comparison of diagnostic ratios of PAHs from A. traffic (past studies), B. Cooking(past studies) and C. this study

A.

	(Gu etal 2010)	(Miguel and Pereira, 1989) (combustion)			Akyüz and Çabuk, 2010		Yunker et al., 2002				Pies et al., 2008		Sofowote et al. (2010)			Zencak et al., 2007.			Wu et al., 2007.		Mantis et al., 2005.
	Shangai	TRAFFIC	PETROLEUM	GRASS	Coal combustio	Vehicular	Petrogenic	Combustion	Petroleum combustion	Grass, wood and coal	Petrogenic	Pyrogenic	diesel exh	urban dust	coal combusti	Coal combustio	Vehicular	Wood combustion	Coal combusti	Vehicular	Wood combusti
Phe/(Phe+Ant)																					
Flu/(Flu+Pyr)	0.52	0.4-.5	<0.2	>0.5												>0.5	0.4-0.5	>0.5			
BaA/(BaA+CHR)	0.27				0.2–0.35	>0.35	<0.2	>0.35					0.2-0.25	0.3-0.35	0.55-0.6						0.43
Ind/(Ind+Bpe)																					
BeP/(BaP+BeP)	0.63																				
Ant/(Ant+Phe)	0.13						<0.1	>0.1			<0.1	>0.1									
IcP/(IcP+BgP)	0.45						<0.2		0.2–0.5	>0.5											
BaP/(BaP+BeP)							0.2-0.4														
BaP/BghiP																			0.9-6.6	0.3-0.44	

B.

	COOKING(See et al(2006)			He et al., 2004		Zhu and Wang, 2003				See and Balasubramanian, 2008				
	CHINESE	MALAY	INDIAN	CHINESE, HUNAN	CHINESE, CANTONESE	CHINESE	CHINESE	CHINESE	CHINESE	STEAMING	BOILING	STIR-FRY	PAN-FRY	DEEP-FRY
	$PM_{2.5}$	$PM_{2.5}$	$PM_{2.5}$	$PM_{2.5}$	$PM_{2.5}$	TSP and gas	TSP and gas	TSP and gas	TSP and gas	$PM_{2.5}$	$PM_{2.5}$	$PM_{2.5}$	$PM_{2.5}$	$PM_{2.5}$
Phe/(Phe+Ant)	0.21	0.28	0.21	0.96	1	0.51	0.41	0.37	0.51	0.96	0.96	0.94	0.94	0.95
Flu/(Flu+Pyr)	0.32	0.38	0.43	0.44	0.36	0.18	0.19	0.23	0.23	0.51	0.52	0.56	0.55	0.53
BaA/(BaA+CHR)	0.4	0.32	0.5	0.51	0.47	0.74	0.18	0.22	0.38	0.31	0.34	0.28	0.29	0.28
Ind/(Ind+Bpe)	0.43	0.38	0.39	–	0.19	–	–	–	–	0.54	0.52	0.45	0.46	0.44

C.

	INDIAN	WESTERN	AFRICAN	CHINESE
BeP/BeP+BaP)	0.08	0.10	0.27	0.31
IND(IND+BghiP)	0.48	0.52	0.58	0.37
Cor/Bep	1.84	10.45	2.31	3.55
BghiP/BeP	10.02	12.31	1.63	2.47

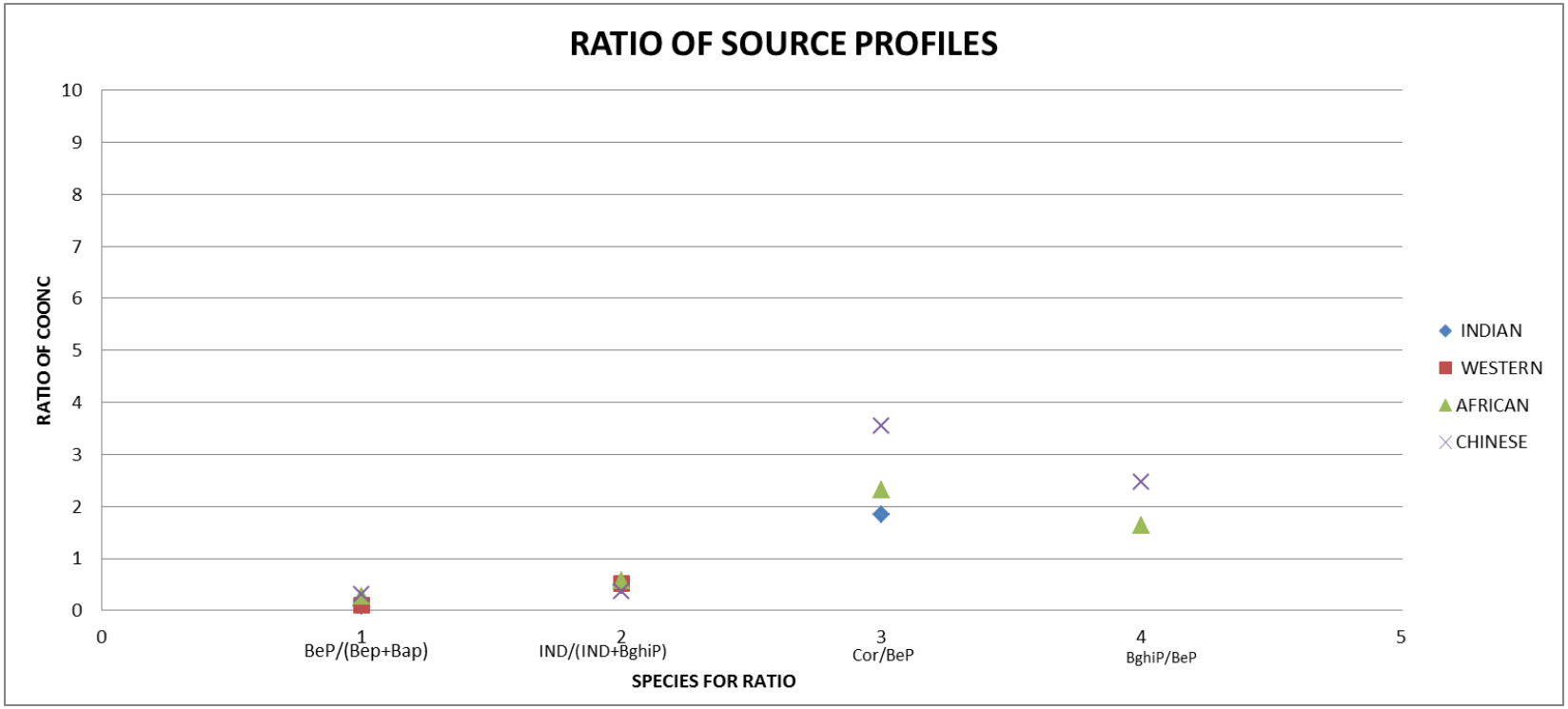


Figure 27 Diagnostic ratio of profiles PAH

Table 35 Diagnostic ratios of acids.

					mg/kg meat	mg/kg meat	ug/kg meat	
using PROFILE(ug/ug OC)	GAS WEST	GAS AFR	GAS CHINE	GAS IND	rogge frying burger	rogge charbroil burger	schauer potatoes	schaur charbroil
oleic/linol	2.08	2.04	0.82	2.34			1.11	6.69
chole/palmitic	0.14	0.05	0.06	0.12	0.50	0.32		0.02
stearic/palmitic	0.17	0.37	0.62	0.35	0.61	0.67	0.48	0.59
oleic/stear	9.57	2.35	2.63	5.99	1.16	1.46	2.29	2.21
using CONCENTRATION (ug/m3)	WESTERN	AFRICAN	CHINESE	INDIAN				
oleic/linol	2.02	2.02	1.58	2.43				
chole/palmitic	0.13	0.03	0.04	0.17				
stearic/palmitic	0.19	0.24	0.43	0.45				
oleic/stear	9.75	4.05	3.58	6.05				

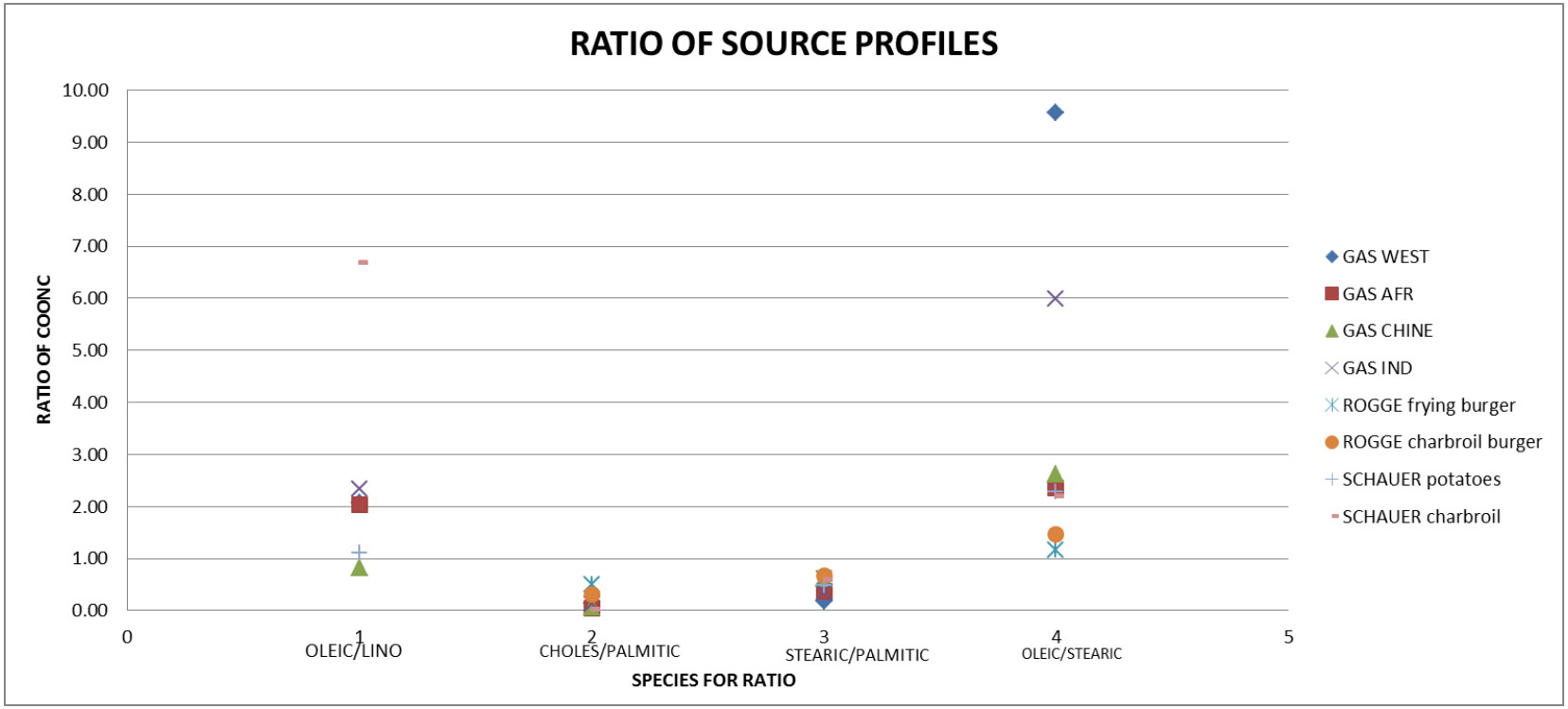


Figure 28 Diagnostic ratio of acids

3.6 Total compounds emitted.

Analysis of the total concentrations across the group of compounds presented in Table 36showed that generally Chinese cooking style emitted the highest concentration of compounds across the entire range of cooking techniques. Acids were the most prominent compounds released during Chinese cooking with total concentration of 21.61µg/m^3. African style cooking had the lowest total concentration when compared to all other styles with a concentration of 0.37µg/m^3 of total sterol emitted compared to1.34 µg/m^3 from Chinese style cooking (which was observed to emit the highest total concentration of sterols). For Indian and western style cooking the most prominent set of compounds released were alkanes whereas similar to observation for Chinese cooking, PAH were the highest total emitted compounds for African cooking (6.83 µg/m^3). High concentrations of monoglycerides were observed in Western and Chinese cooking, 10.33µg/$m^3$and 11.52µg/m^3 respectively. Table 36 and the pie chart below represent the average concentrations emitted from cooking styles.

Figure 30 shows pie charts for total concentration of compounds emitted from cooking from Western, Chinese and Hunan style cooking (A,B,C respectively). For Chinese cooking by Zhao et al., 2007 highest total concentration emitted were for alkanes and PAH collectively accounting for about 75% of the total concentration. For the western style cooking a larger percentage was represented by alkanes than in Chinese cooking and a smaller fraction for PAH. In these studies the percentage for acid is much less than in the present study. However for the hunan style cooking in (C) high concentration of acid, a lot higher fraction than was reported by He et al., 2004.

Table 36 Total concentrations of compounds (alkane, PAH, sterol, glyceride and acids) at cooking source (µg/m^3)

(µg/m3)	INDIAN	WESTERN	AFRICAN	CHINESE
Total n-ALKANES	12.41	11.66	4.67	12.99
Total PAH	5.35	9.31	2.92	12.74
Total ACID	6.65	9.87	6.83	21.61
Total STEROLS	1.18	0.94	0.37	1.34
Total MONOGLYCERIDE	3.38	10.33	1.48	11.52

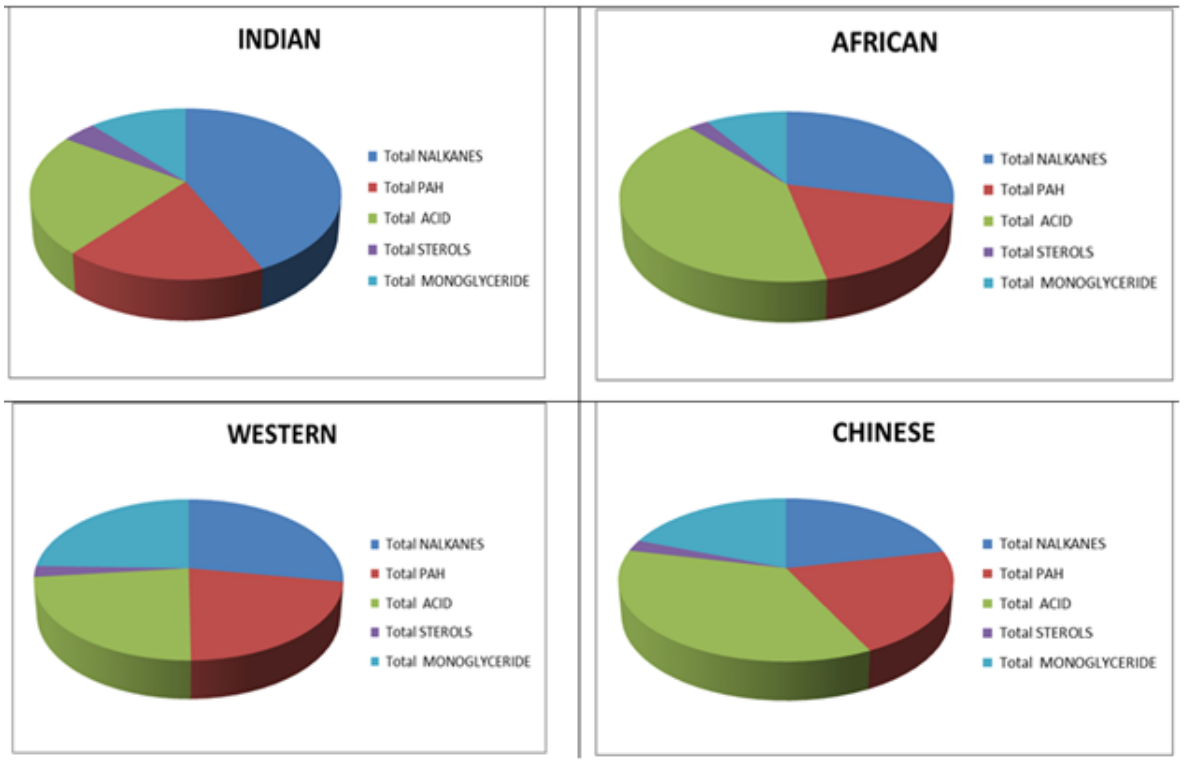
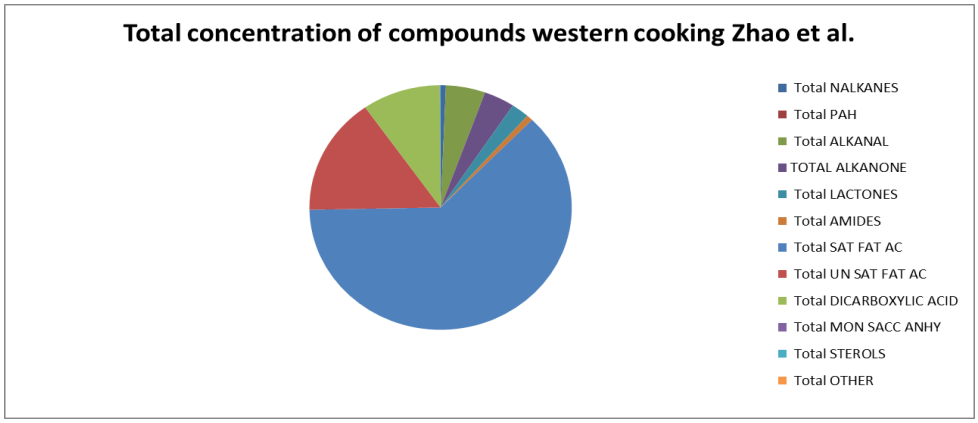
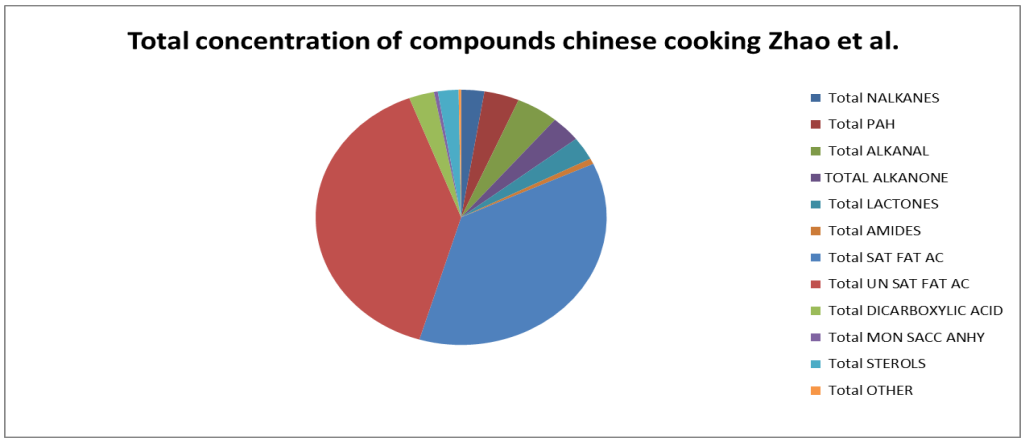


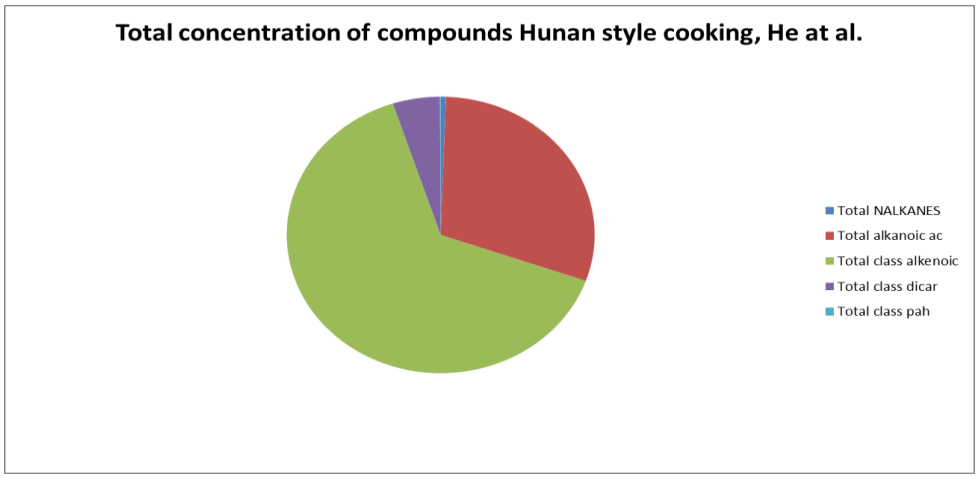
Figure 29 Pie chart of total concentration of compounds from cooking source (µg/m^3)



A. TOTAL CONCENTRATION OF COMPOUNDS –WESTERN COOKING PROFILE ZHAO ET AL., 2007.



B. TOTAL CONCENTRATION OF COMPOUNDS –CHINESE COOKING PROFILE ZHAO ET AL., 2007.



C. TOTAL CONCENTRATION OF COMPOUNDS –HUNAN COOKING He et Al

Figure 30 He et al., 2004 and Zhao et al., 2007 comparism of total concentration of compounds emitted

Figure 30 shows pie charts for total concentration of compounds emitted from cooking from Western, Chinese and Hunan style cooking (A,B,C respectively). For Chinese cooking by Zhao et al., 2007, highest total concentrations emitted were for alkanes and PAH collectively accounting for about 75% of the total concentration. For the western style cooking a larger percentage was represented by alkanes than in Chinese cooking and a smaller fraction for PAH. In these studies the percentage for acid is much less than in the present study. However for the hunan style cooking in (C) high concentration of acid, a lot higher fraction than was reported by He et al., 2004.

3.7 Cooking profiles

Individual source profiles were made up of the average and standard deviation of species abundances for all individual profiles within a group. Source profiles in this case were prepared with respect to OC for cooking with gas source as this was identified as the most frequently used heat source in restaurants.

Below Table 37 is a list of cooking profiles µg/ µg of OC.

Table 37 Source profile of cooking µg/ µg of OC –with Gas.

	WEST	ARF	CHI	IND		WEST	ARF	CHI	IND
EC	2.66E-03	1.21E-03	3.19E-03	3.79E-03	DODE	7.49E-03	3.06E-03	5.46E-03	9.45E-03
ECU	1.00E-08	1.00E-08	1.00E-08	1.00E-08	DODEU	5.61E-03	2.51E-03	6.83E-03	3.18E-03
LEVOG	1.17E-02	6.14E-03	5.25E-03	1.75E-02	NONDIA	2.28E-03	1.01E-02	2.18E-03	2.03E-03
LEVOGU	6.26E-03	4.08E-03	2.67E-03	3.62E-03	NONDIAU	1.25E-03	1.33E-02	2.03E-03	1.96E-03
CHOL	2.30E-03	1.17E-03	8.02E-04	2.47E-03	TRI	6.10E-03	3.65E-04	1.27E-04	1.53E-03
CHOLU	3.18E-04	4.09E-02	2.37E-02	5.90E-02	TRIU	9.54E-03	3.12E-04	1.14E-04	8.46E-04
PICENE	2.56E-03	1.09E-03	1.29E-03	2.27E-03	TETDE	9.69E-03	9.43E-04	1.61E-03	3.32E-03
PICENEU	1.61E-03	1.26E-03	4.18E-04	2.72E-03	TETDEU	1.46E-02	1.16E-03	1.72E-03	2.43E-03
BZBFLU	2.17E-03	6.29E-04	5.68E-04	8.90E-04	PENT	6.07E-03	2.81E-03	1.81E-03	7.90E-03
BZBFLUU	1.46E-03	5.79E-04	2.62E-04	7.93E-04	PENTU	4.03E-03	2.40E-03	1.07E-03	3.71E-03
BZKFLU	1.57E-04	2.48E-04	2.15E-04	5.25E-04	HEP	5.57E-04	2.32E-04	1.45E-04	1.85E-04
BZKFLUU	6.82E-05	1.92E-04	3.08E-04	4.91E-04	HEPU	7.17E-04	1.89E-04	6.97E-05	6.10E-05
BZEPYR	1.58E-04	4.02E-04	4.55E-04	1.43E-04	NONA	2.49E-04	1.75E-04	1.79E-04	2.88E-04
BZEPYRU	3.08E-04	5.97E-04	4.65E-04	2.47E-04	NONAU	4.59E-05	1.06E-04	1.51E-04	2.29E-04
INDPYR	1.98E-03	5.87E-04	5.68E-04	1.12E-03	EICO	2.09E-04	2.94E-04	4.13E-04	3.75E-04
INDPYRU	1.49E-03	3.82E-04	1.91E-04	8.74E-04	EICOU	1.92E-04	3.59E-04	2.96E-04	1.85E-04
BZGHPL	1.68E-03	4.95E-04	1.05E-03	1.24E-03	DOCO	6.01E-03	5.13E-04	4.70E-04	5.07E-03
BZGHPLU	1.22E-03	3.58E-04	5.30E-04	1.04E-03	DOCOU	6.94E-03	5.40E-04	1.49E-04	5.53E-03
PALMTA	1.70E-02	2.59E-02	1.40E-02	1.99E-02	TETCO	1.04E-03	4.10E-04	3.96E-04	8.35E-04
PALMTAU	1.88E-02	3.77E-02	2.33E-02	3.92E-02	TETCOU	4.30E-04	2.19E-04	3.73E-05	2.56E-04
LINOLA	1.32E-02	1.12E-02	2.75E-02	1.77E-02	MONMY	3.71E-02	6.10E-03	1.23E-02	1.59E-02
LINOLAU	1.53E-02	1.47E-02	4.87E-02	2.35E-02	MONMYU	2.21E-02	5.79E-03	1.29E-02	9.00E-03
OLA	2.75E-02	2.28E-02	2.27E-02	4.14E-02	MONPA	4.46E-02	4.71E-03	2.79E-02	1.24E-02
OLAU	2.44E-02	3.03E-02	2.61E-02	4.03E-02	MONPAU	4.60E-02	3.69E-03	4.40E-02	7.02E-03
STEARA	2.87E-03	9.70E-03	8.64E-03	6.91E-03	MONOL	4.42E-02	1.21E-02	2.42E-02	1.48E-02
STEARAU	3.08E-03	7.35E-03	9.78E-03	7.57E-03	MONOLU	3.64E-02	1.38E-02	3.72E-02	1.07E-02
UNDEC	2.30E-02	1.71E-03	1.35E-03	4.70E-03	MONSTE	2.31E-02	9.50E-03	1.51E-02	1.58E-02
UNDECU	2.56E-02	1.26E-03	6.75E-04	5.17E-03	MONSTEU	5.10E-03	9.33E-03	1.71E-02	8.97E-03
OCTA	1.28E-02	3.49E-03	5.37E-03	2.12E-03					
OCTAU	1.20E-02	5.52E-03	7.42E-03	1.39E-03					

COMPARABILITY AMONG THE SOURCE PROFILES

Kong et al 2011 have successfully used a self-normalizing statistic called Coefficient of divergence (CD) to calculate the degree of similarity between source profiles using the following formula (Wongphatarakul al., 1998) even though the formula was formally only used to understand the degree of similarity between PM observations across different sites (Wongphatarakul al., 1998). CD values range from 0 to 1 with values closer to zero indicating a higher degree of similarity while values closer to one indicating concentrations are different.

$$CD_{jk} = \sqrt{\frac{1}{p}\sum_{d=1}^{p}\left(\frac{x_{ij} - x_{ik}}{x_{ij} + x_{ik}}\right)^2}$$

Where

CD is the coefficient of divergence

xij is the concentration of species i in profile j

xik is the concentration of species i in profile k

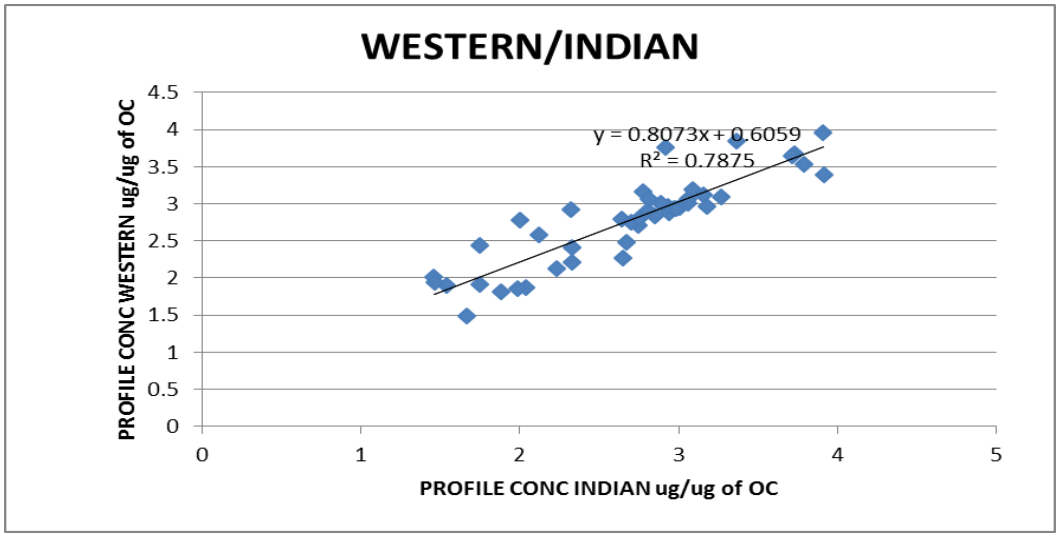
p is the number of species investigated.

Using the profiles in the table above the Coefficient of Divergence were calculated and presented in Table 38 below which shows that the highest similarities in profiles are observed between Chinese and African profile with a CD of 0.27. African and Western profiles are the profiles that are least similar with a CD value of 0.47.

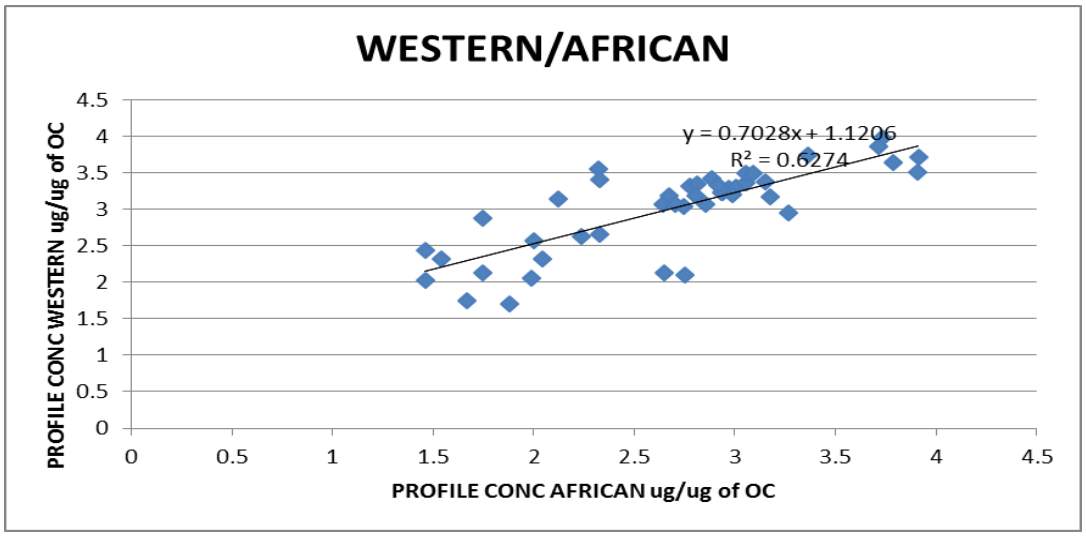
Table 38 Coefficient of divergence for cooking profiles

	Western	African	Indian	Chinese
west	0	0.47	0.31	0.45
african		0	0.39	0.27
indian			0	0.41
chinese				0

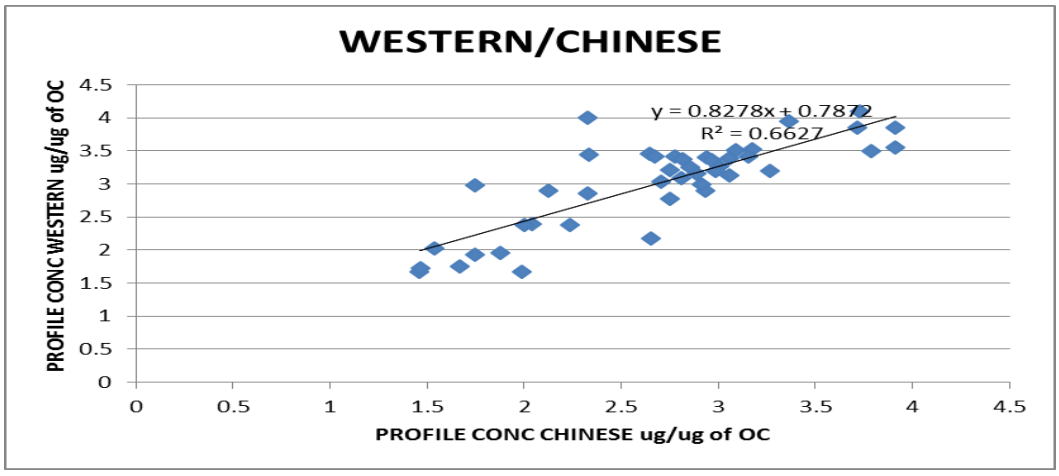
Plots of the source profiles in µg/µg of organic carbon (OC) are made and are shown in Figure 31 (a-f). The values of r^2 range from (0.62- 0.85). The highest r^2 value was found for the African and Chinese regression (f) with a value of 0.85. R^2 value is lowest for Western and African profiles when compared with each other (0.62). Thus a goodness of fit exists for the African and Chinese profiles when compared with each other and are thus the most similar.



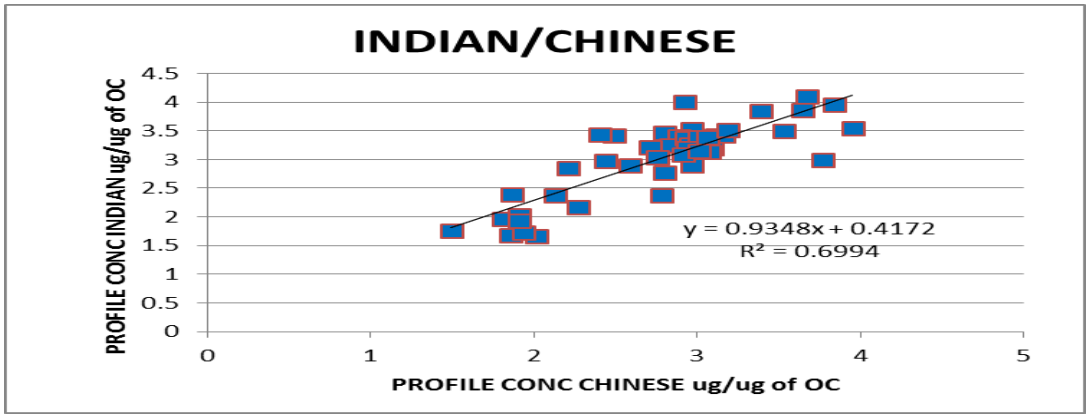
a. Western /Indian



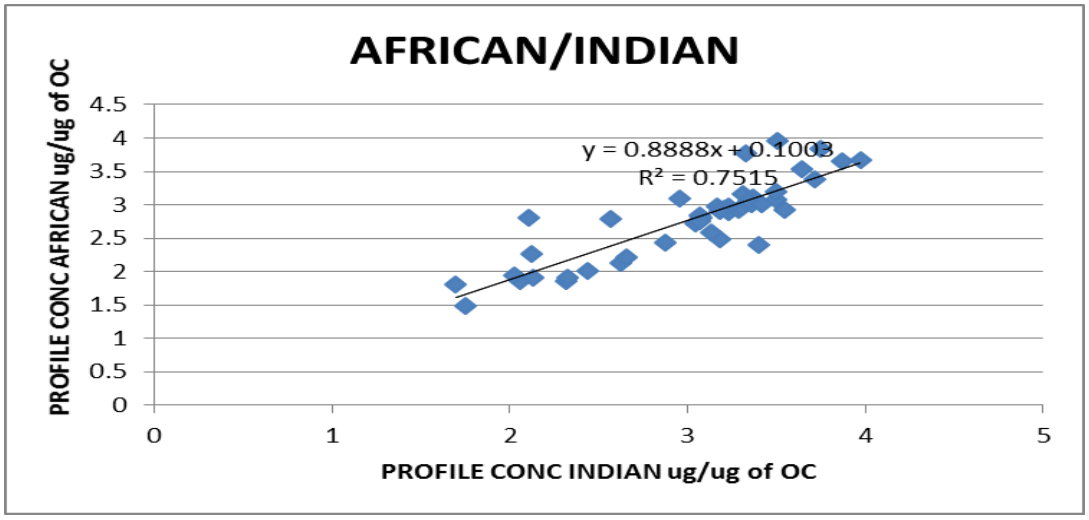
b. Western/African



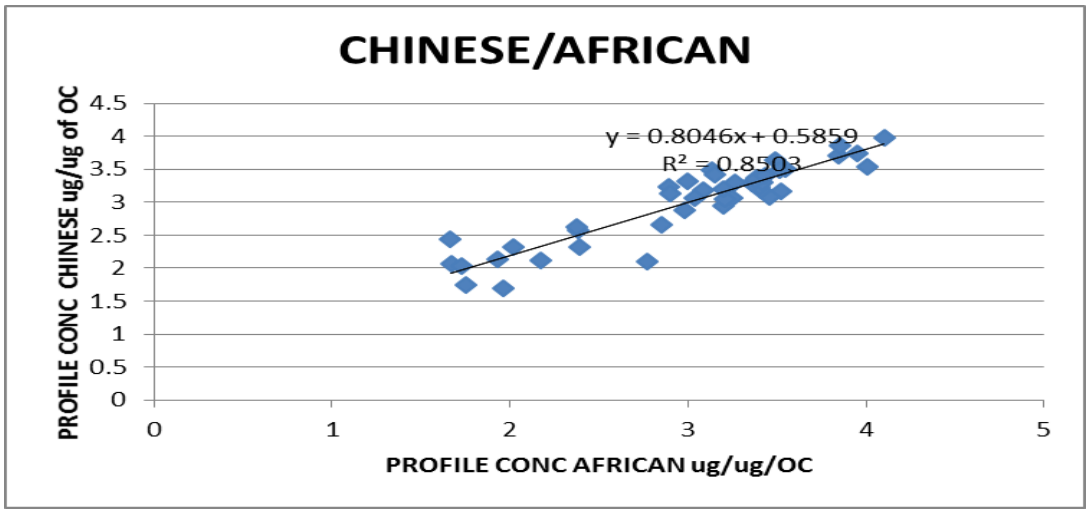
c. Western/Chinese



d. Indian/ Chinese



e. African /Indian



f. Chinese/ African

Figure 31 Plots of cooking profiles (µg/µg of OC) against each other (with R^2 values)

An analysis of the profiles of Western style and Chinese style cooking against themselves for a study by Zhao et al., 2007c(

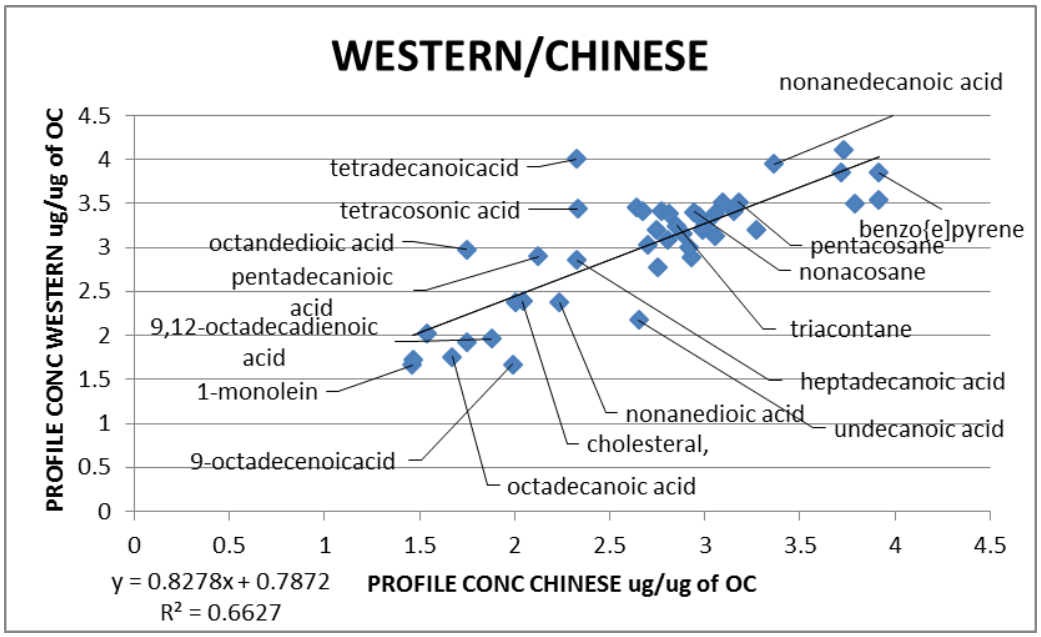


Figure **32** A) and the present study (

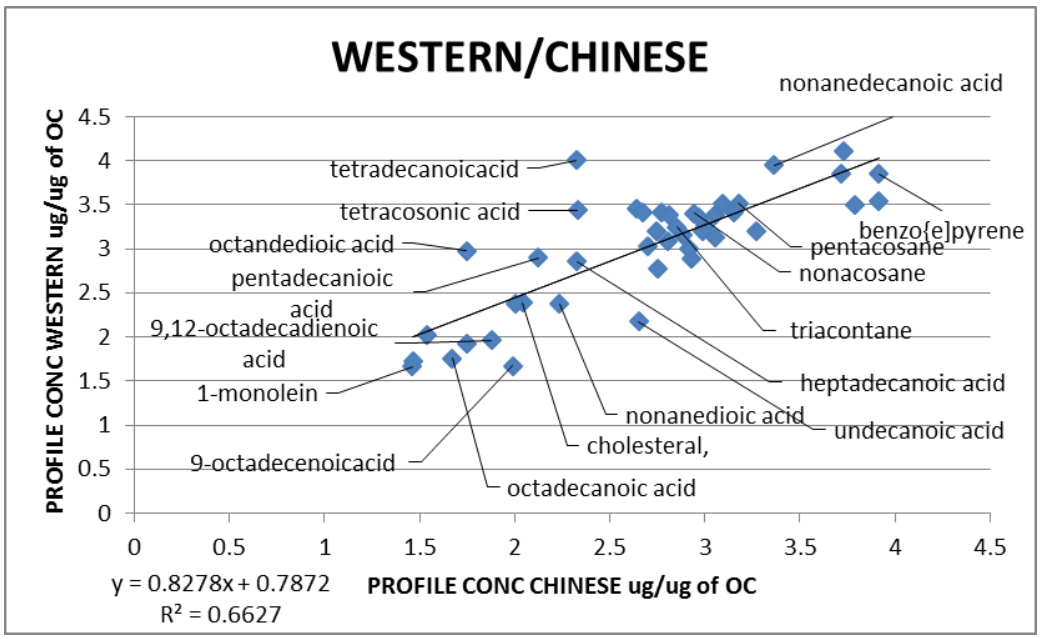


Figure **32** B) present plots that show a better correlation between the two cooking styles in this study than previously observed by Zhao. Higher correlation is seen between the two profiles especially for alkanes, acids and glycerides. In

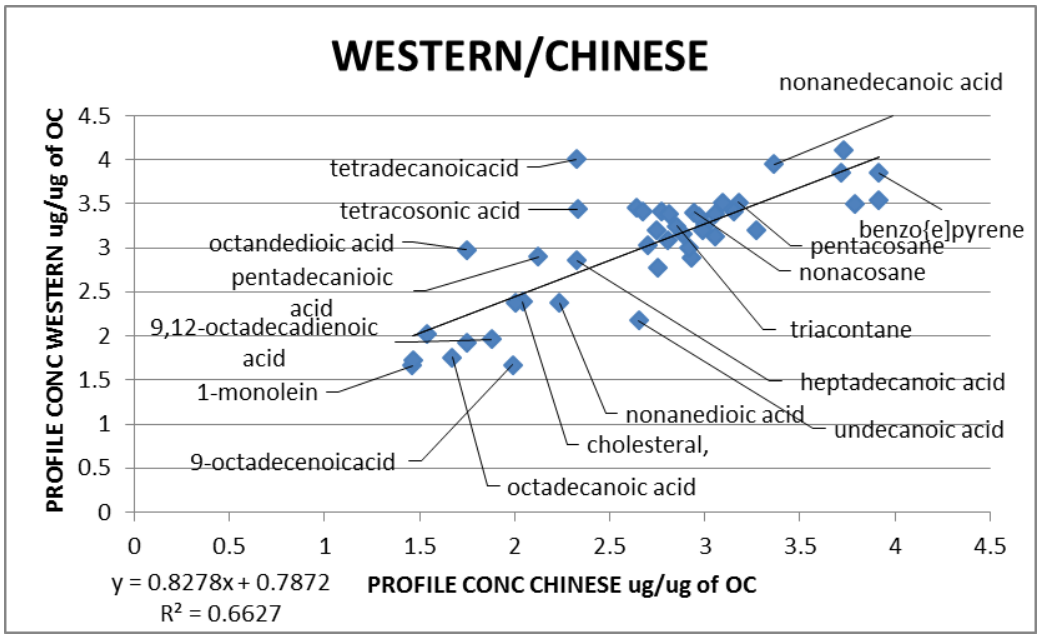


Figure **32**B, both profiles are found to have similar concentration of acids as such these can serve as good markers for cooking (hexadecanoic acid, 9,12-octadecadienoic, 1-monoolein, 1-monostearin). 9-octadecanoic acid acid and tetradecanoic acid is seen to lie away from the trendline for the cooking profiles so may not be a good representative for both profiles.

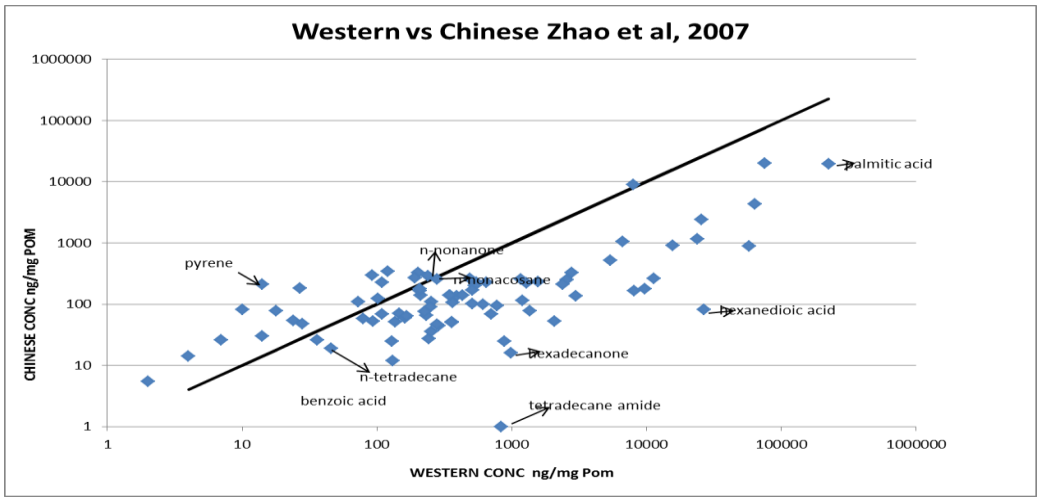
ANOVA- Analysis of variance was carried out for the cooking profiles

	MEAN SQ	SUMMARY OF SQ	df
AFRICAN	0.167	7.18	43
INDIAN	0.31	13.3	43

CHINESE	1.81	77.9	43
WESTERN	0.53	25.7	43

Using ANOVA- There was a statistically significant difference between groups as determined by one-way ANOVA $p = .038$. A Tukey post hoc test revealed that the concentration emitted from African cooking was statistically significantly lower than Indian cooking and western style cooking with Chinese cooking having the highest mean concentrations.

A. Zhao et al, 2007c



B. This study

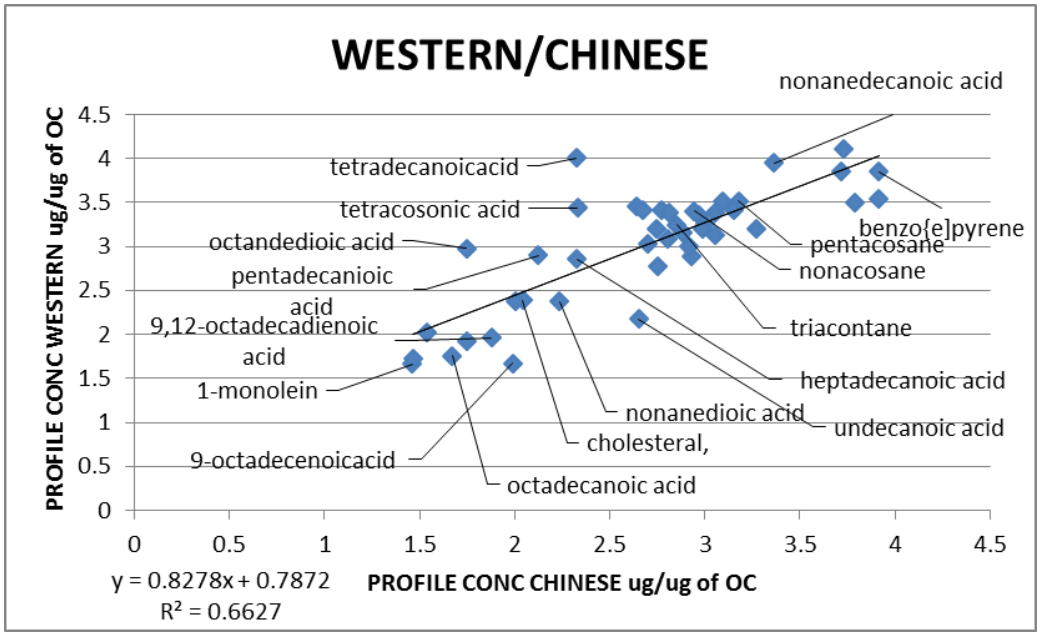


Figure 32 Analysis of profile for Chinese and Western cooking in A. China by Zhao et el, 2007c; and B. in this study.

3.8 Discussion of AMS Results from Manchester Birmingham campaign

The measurements for this study were conducted as part of a Project between the University of Birmingham and University of Manchester (Cook off project). The aim of the sampling was to understand clearly the signature output for cooking from the Aerosol mass Spectrophotometer.

Sampling was carried out between 20th March 2012 and 22nd March 2012 where foods were cooked in the designed laboratory kitchen described in chapter 2**.** Different types of oils were fried in glass beads to simulate the cooking process and emissions only associated to oils. During these cooking experiments, the Aerosol mass Spectrophotometer, operated by D.E. Young (from the University of Manchester), was taking samples in the exhaust duct to analyse for the organic loading during cooking. Data of the AMS has been provided by D.E. Young, and is displayed in Figure 33, Figure 34, Figure 35 and Figure 36. While the AMS samples

were being collected, PM samples were collected from the duct of the trailer on Teflon filters for gravimetric analysis.

Below in Table 39 is a list of foods that were cooked and the time the cooking took place during the sampling. In between each cooking experiment the laboratory kitchen was ventilated.

From the output of the AMS, the non-refractory mass concentrations during the sampling period generally show that organics are the dominant fraction.

On day 1 it was observed that mass loading was high especially during the periods that meat was being grilled, followed by the grilling of vegetables. Grilling of sea food was found to emit the least amount of organic. Generally grilling was found to lead to the largest quantity of organics with concentration of about 4500 $\mu g/m^3$.

On day 2 the shallow frying of chicken was found to emit the largest mass loading in the AMS with less emissions during deep frying of chicken, chips and samosa and the lowest observed concentration was observed during deep frying of fish and chips.

Stir frying of food was observed to result in the release of high organics especially for Chinese stir frying with chicken, less concentration was observed for stir frying with sea food even though the loading was quite significantly high. Lower concentration was observed for Indian stir fry/stewing method. This high concentration can be attributed to the physical stirring of the ingredients during grilling and frying found to lead aerosols due to the process of splashing of the ingredients (Long et al., 2000).

Analysis of all the AMS data for the sampling days showed that grilling and stir frying of meat and chicken lead to the largest quantity of organics (~ 4500 $\mu g/m^3$). Stewing of tikka masala emitted less organics with a maximum quantity of 900 $\mu g/m^3$, with the least amount of organic loading for deep frying (fish and chips were as low as 320 $\mu g/m^3$ while chicken frying had a maximum load of 600 $\mu g/m^3$). It was interesting to find that grilling of seafood lead to lower

mass loading (1000 µg/m^3) than other grilling processes involving ingredients like meat and vegetables.

It was observed in Figure 33 that during grilling the diameter representing the highest particle number concentration were within the range of the diameter 35nm-60nm which was within the range of diameters observed in previous studies in Table 6 where the range of particles generated from cooking were found to lie between 20-100nm. Buonanno et al in 2011 found the particle diameter mode for grilling of bacon to be 50nm and eggplant 40nm. In this study grilling of meat was found to produce particles of 40nm while particles generated during seafood grilling were about 35nm in size with larger particles of about 60nm diameter emitted during grilling of vegetable. Larger concentrations of particles were emitted during the grilling of meat, $2.5x10^6$particles/cm^3 as compared to $1.6x10^6$particles/cm^3 and $1.26x10^6$particles/$cm^3$for grilling of seafood and vegetables respectively.

In Figure 34 a much smaller particle size was emitted during frying with particles of diameter between 15 to 25nm generated during this cooking method. Larger particle sizes were generated during shallow frying than when deep frying. It was noted that for shallow frying, grilling and stir frying the particles generated were of similar size of about 25nm diameter, this could be due to the similar process whereby the ingredients are exposed to heat directly and so undergo similar breakdown procedures. The largest particle size from analysis of the data was generated during the grilling of vegetables. The concentration of particles was much higher on day 2 during frying with the shallow frying of vegetables and chicken having the highest value of $9x10^6$particles//cm^3, deep frying of fish and chips $4x10^6$particles/$cm^3$and deep frying of plantain and samosa and chicken $6x10^6$particles/cm^3. These findings are consistent to what was observed in studies by Wallace et al., 2004, Buonanno et al., 2011, Huboyo et al., 2011a, Hussein et al., 2006 where higher concentration of particles were generated for frying than all cooking methods as discussed earlier in Section 1.6.

On day 3 the stir frying of seafood and chicken generated similar size particles of about 25nm diameter as seen in Figure 35. Stir frying/ stew of Indian tikka masala generated mainly particles of about 15nm and the concentration was lower than other cooking methods with about $450x10^3$particles/cm^3. The concentration for stir frying of chicken and seafood were $2.5x10^6$particles/cm^3 and $3.5x10^6$particles/cm^3.

Similar to observations by previous studies the lowest particle number concentration was observed for cooking activities involving water (Indian stewing with boiled rice) and highest concentration for frying however shallow frying of chicken and vegetables emitted more particles than deep frying in this study slightly different from what was observed by See and Balasubramanian in 2006. In their study deep frying produced higher number concentration than pan frying ($59x10^4$particles/cm^3 and $11x10^4$particles/cm^3 respectively)

Figure 37 shows the gravimetric weight obtained from filter exposure during the various cooking experiments. It was observed that the concentration trend are similar to that from the SMPS with high concentrations observed during grilling of meat(1000 µg/m^3) and shallow frying of chicken eggs and vegetables (750µg/m^3). From the filters stir frying generally resulted higher PM concentration (750-320 µg/m^3) than deep frying(110-80 µg/m^3). The stewing of Indian tikka masala with rice was observed to have emitted similar concentration with deep frying and stew food grilling of 100 µg/m^3.

Figure 36 shows AMS and SMPS data obtained from the frying of different cooking oils in glass beads. Supermarket vegetable oil and sunflower oil are observed to have similar concentrations and particle mode diameters, on the SMPS the total number count was $4x10^6$particles/cm^3 and $2.5x10^6$particles/cm^3 respectively with mode diameter of 50nm with AMS concentration of 12 $x10^3$µg/m^3 for both oils. Chinese stir fry oil and olive oil had mode at 30nm and another peak at 100nm with concentration of $16x10^3$particles/cm^3 and $20x10^3$particles/cm^3. Olive oil was found to have the highest mass loading concentration

among the oils analysed with a peak concentration of 1.4 x10^3µg/m^3. Previously in cooking studies (Table 6), particle mode diameters correspond with what was observed in this study. Yeung and To, 2008 reported a mode diameter of 107nm for hot oil analysis, Siegmann and Sattler, 1996 fried rapeseed oil and obtained a mode diameter between30-100 while Glytsos et al., 2010 fried a slice of onion in olive oil and the mode diameter found was 20-45nm.

A study to characterize indoor sources of particles conducted in Boston, USA, made measurements of particle size and volume concentration over 6 days in four non-smoking households equipped with gas and electric stoves (Abt et al., 2000). It was found that the highest mean peak mass concentrations were for barbequing and sautéing for the $PM_{0.02-0.5}$ and $PM_{0.7-10}$ respectively, whilst the lower mean peak concentrations were found for frying and oven cooking or toasting for the same size ranges respectively (Abt et al., 2000). When comparism is made with the present study similar size particles was obtained for grilling as for the barbeque.

Table 39 Schedule of cooking activities during COOK OFF experiment.

	MORNING	VENTILATION 1 HOUR	AFTERNOONa	VENTILATION ONE HOUR	AFTERNOONb
Grilling 20/3/2012	meat		Sea food		vegetables

Frying **21/3/2012**	Deep fry chips and fish		Deep fry plaintain, chicken and pastries		Shallow fry chicken and vegetables
Stir fry and stew 22/3/2012	Stir fry sea food with fried rice		Stir fried meat and fried rice		Chicken tikka masala and rice

Day 1 grilling

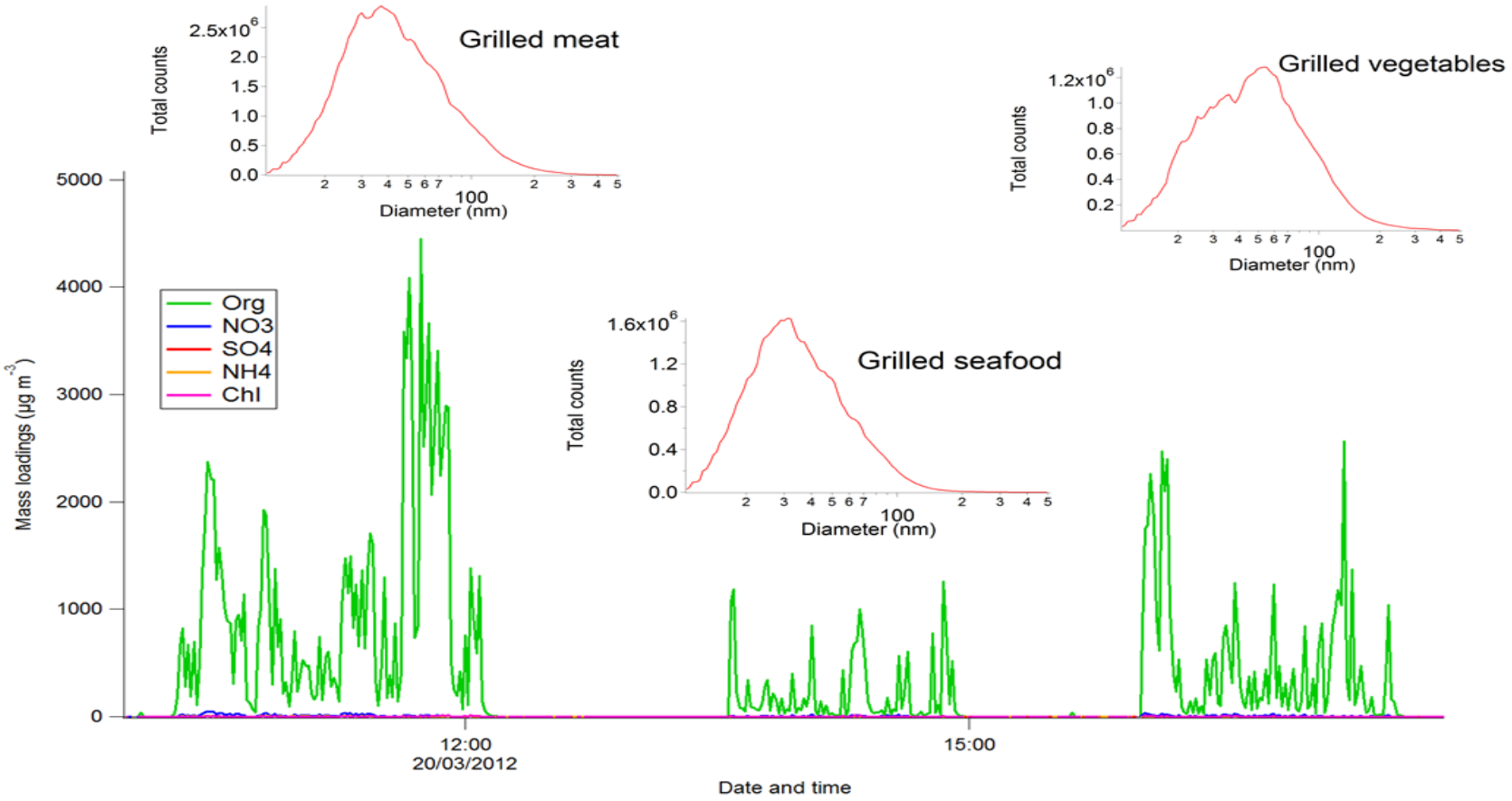


Figure 33 AMS and SMPS data on day 1- GRILLING of meat, seafood and vegetables. Data provided by D.E. Young.

Day 2 deep and shallow fry

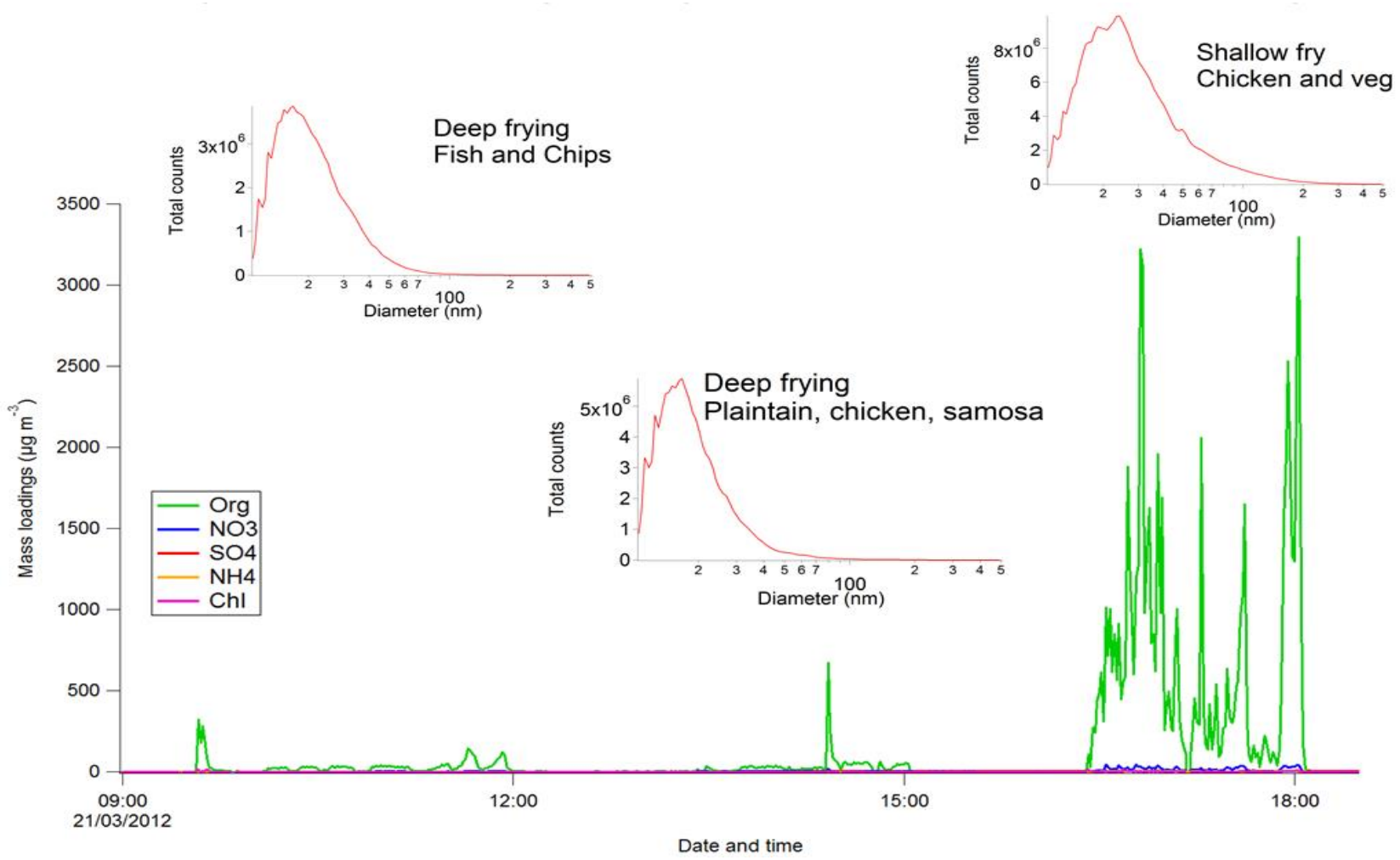


Figure 34 AMS and SMPS data on day 2- FRYING –deep frying and shallow frying. Data provided by D.E. Young.

Day3- Stir fry

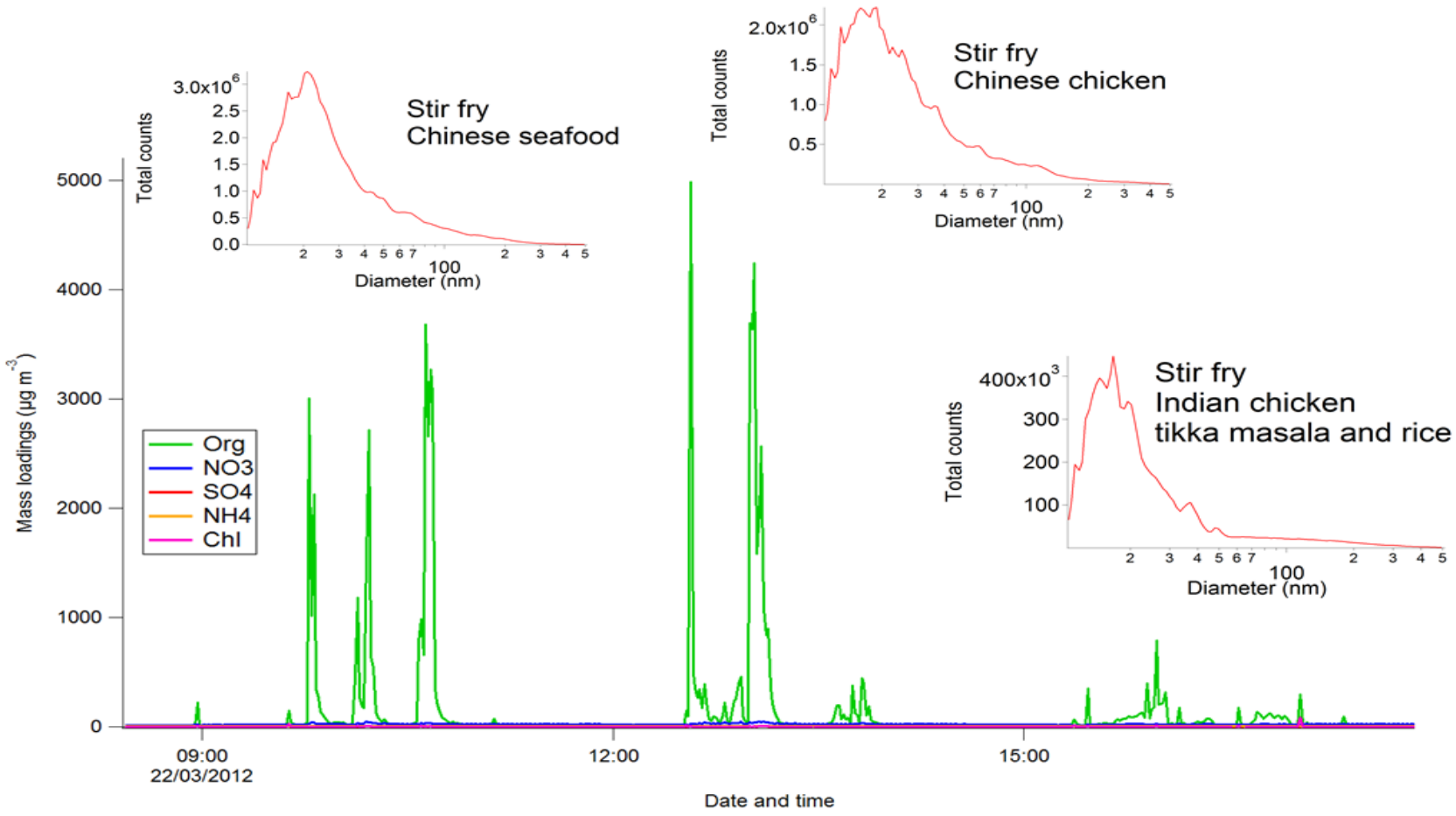


Figure 35 AMS and SMPS data on day 3- STIR FRY of seafood, chicken and STEWING. Data provided by D.E. Young.

Day3 B- Oils

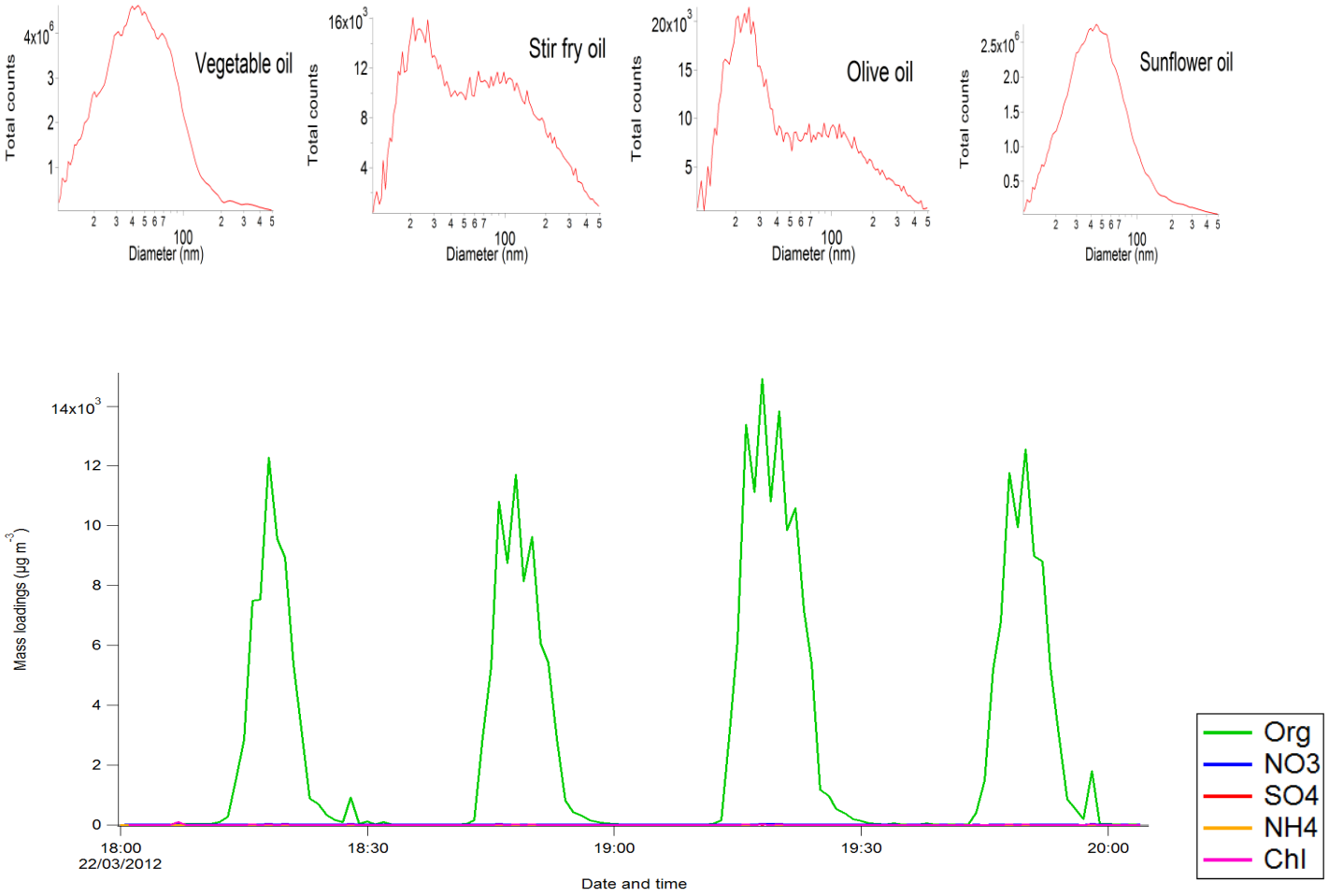


Figure 36 AMS and SMPS data on day 3- FRYING OF OIL IN GLASS BEADS. Data provided by D.E. Young.

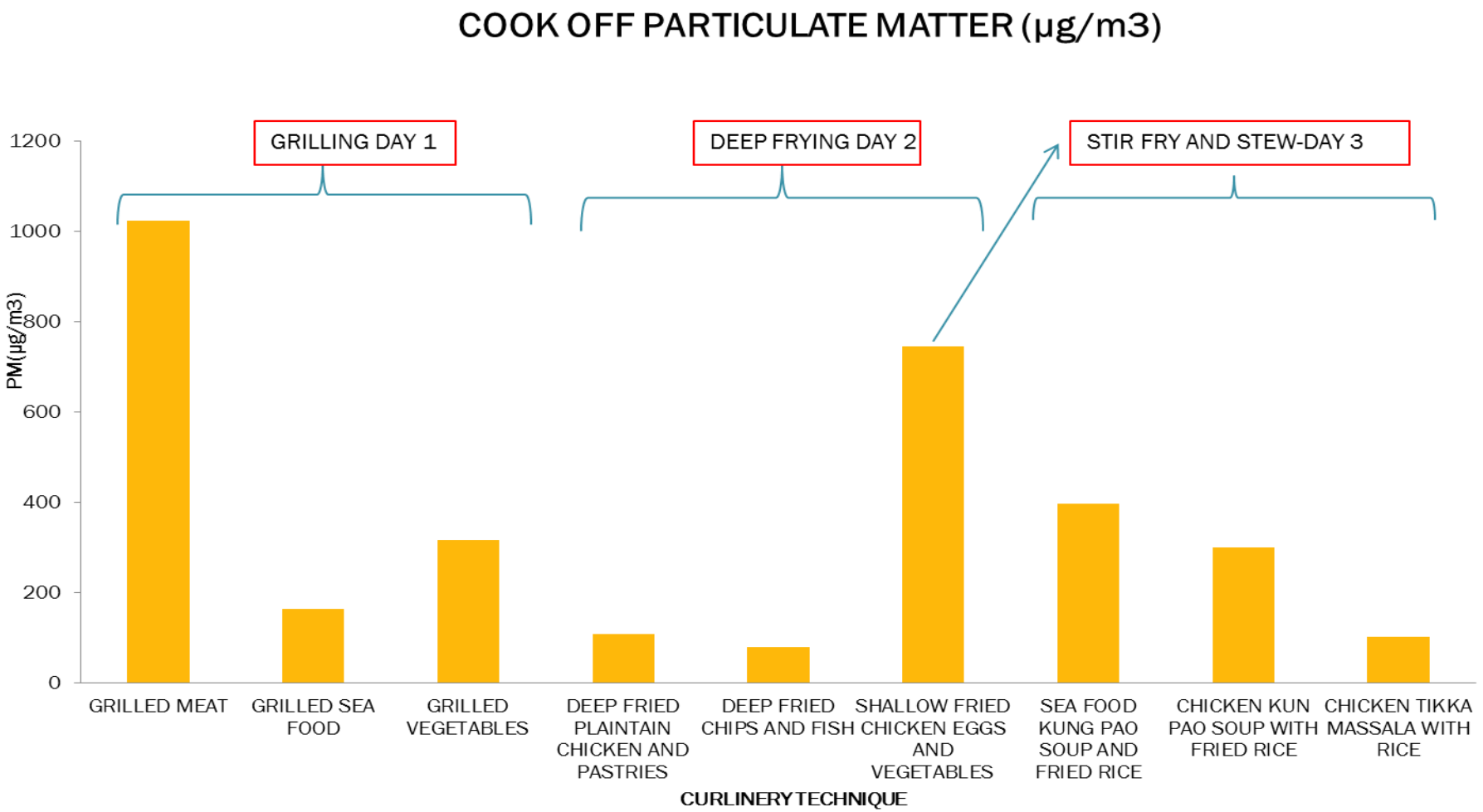


Figure 37 Gravimetric concentration of PM during Cook Off.

3.9 Conclusion

PM samples were collected and analysed in a designed trailer kitchen located in the University of Birmingham while cooking of chicken and rice and potatoes was carried out using different styles of cooking and different fuel sources. The samples were analysed to determine the gravimetric contribution to Particulate matter from the various cooking styles.

It was observed that electric cooking generally resulted in higher concentrations of Particulate matter than when cooking with gas. Higher concentrations of PM were emitted when cooking using the Chinese curlinary technique.

The samples were analysed for their organic composition with Acids being the most prominent compounds released during Chinese cooking with total concentration of 21.61µg/m^3. African style cooking had the lowest total concentration when compared to all other styles with a concentration of 0.37µg/m^3 of total sterol emitted compared to1.34 µg/m^3 from Chinese style cooking. The main group of compounds released during Indian and western style alkanes with PAH having the highest concentration when the total emitted compounds for African cooking was analysed (6.83 µg/m^3). High concentrations of monoglycerides were observed in Western and Chinese cooking, 10.33µg/$m^3$and 11.52µg/m^3 respectively.

The particulate matter mitted from cooking were analyse with an AMS and it was found that grilling and stir frying of meat and chicken lead to the largest quantity of organics (~ 4500 µg/m^3). Stewing of tikka masala emitted less organics with a maximum quantity of 900 µg/m^3, with the least amount of organic loading for deep frying (fish and chips were as low as 320 µg/m^3 while chicken frying had a maximum load of 600 µg/m^3). This signified that high temperature and direct exposure to ingredients to heat in shallow frying can lead to a greater degradation of food ingredients and larger particle generation. It was also found that lowest particle number concentration was observed for cooking activities involving water (Indian stewing with boiled rice) and highest concentration for frying with shallow frying of chicken and vegetables emitting more particles than deep frying.

With all these observations it can be concluded that generally Chinese cooking results in higher concentration of PM and this can be highly attributed to the fact that a lot of shallow frying is involved for the curlinary technique.

CHAPTER 4-Real kitchen sampling.

This chapter presents concentration of particulate matter collected from cooking in a kitchen located in a real home. The aim of the sampling was to quantify the emissions from various cooking styles in a real domestic kitchen.

This chapter contains some sections of verbatim text adapted from the following review article published as part of this PhD:
Abdullahi, L, Delgado Saborit, JM & Harrison, RM 2013, 'Emissions and indoor concentrations of particulate matter and its specific chemical components from cooking: A review' **Atmospheric Environment**, vol 71, pp. 260- 294.

4.1 Introduction

It has been found that a significant part of human exposure to PM occurs indoors as that is where people are found to spend most of their time (Klepeis et al., 2001, Jenkins et al., 1992, Hasheminassab et al., 2014). Up to 80% of people's time is spent indoors (with people in USA and Germany found to spent up to86% of their time indoors) resulting in a lot of emphasis being placed on the understanding of indoor activities that generate PM. With cooking being identified as one of the important sources of $PM_{2.5}$ indoors (Wallace, 2006, Buonanno et al., 2013, Gao et al., 2015, Wan et al., 2011), it is essential to understand its composition, behaviour and how it relates to personal exposure. Measurements taken in a real life kitchen will give an idea of concentrations of compounds emitted during cooking . Several researchers have tried to analyse such concentrations with measurements taken from residential kitchens(Morawska et al., 2003, He et al., 2004b, He et al., 2004a) and commercial restaurants(Lee et al., 2001b, See and Balasubramanian, 2006b). Generally these measurements have been made in countries such as Norway, USA, China, Hong Kong, Japan, Italy, Singapore and Australia and have analysed only a specific type of cooking in each experiment.

A range of organic and inorganic compounds have been found to be emitted during cooking processes; some of which have been identified as possible carcinogens (PAHs) (Abdullahi et al., 2013, See and Balasubramanian, 2006b, Li et al., 1994). High concentrations of these compounds can cause harm to people exposed to them such as cooks and other individuals exposed to the cooking fumes (occupants of the buildings such as children waiting by their parent while meals are prepared, or individuals watching TV in an open plan living room, or the old grandparent keeping warm in the room adjacent to the kitchen). Respiratory diseases have been positively linked with exposure to cooking generated particles (Wang et al., 1996, Gao et al., 1987, Koo and Ho, 1996) with a three-fold increase observed for risk of lung cancer in women with increased number of meals cooked per day (Ko et al., 2000). Ko et al (2000) found that the risk for lung cancer was higher in women that would wait for cooking oil to emit fumes before they started cooking.

4.2 Methodology

The sampling took place between July and August 2014 and in October 2014. Sampling was carried out in the kitchen of a residential house in Birmingham, with no other activities occurring during the cooking exercise in order to minimize the influence of emission from other indoor PM sources. The kitchen was about 9 meters by 4 meters and had a four-burner gas stove connected to the city gas supply system. The sampling instrument was placed on an elevated platform, as shown in **Figure 8**, with its port facing the burner (located 0.5 m from the pan and 1.5 m above the ground). Sampling was carried out with all the windows and door closed. Apart from the investigator, no other person was in the house during the course of the sampling. The kitchen was ventilated between each cooking experiment by opening the window. The cooking duration was about 40-70 minutes, with samples collected via two $PM_{2.5}$ partisol inlets that operated at 16L/min (1 with Teflon filter- for gravimetric analysis and 1with a quartz fibre filter-for organic analysis). After the sampling campaign the filters were stored

in air tight metal tins and placed in the freezer at -22^{o}C. The same type of meals prepared in the trailer were prepared in the real kitchen.

4.2.1 Analytical method

The Teflon filters collected were weighed before and after sampling for particulate mass concentrations using a Sartorius model MC5 microbalance, with a measurement limit of ±1 µg. Prior to sampling all Teflon filters were equilibrated for 24 hours in the weighing room which had a relative humidity of around 35% and a temperature of 20 ± 2 °C, the filters were then weighed and labelled (Yin et al., 2010). After sampling the filters were weighed in the weighing room without 24hr exposure to prevent loss of volatile species collected from the sampling.

The quartz fibre filter samples were analysed for organic species as described in chapter 2. Organic and elemental carbon analysis were carried out using the Sunset Laboratory Thermal-Optical Carbon Aerosol Analyzer as described in chapter 2.

4.3 Gravimetric concentration of emission from cooking in kitchen

The gravimetric concentrations measured from cooking in the kitchen are shown in Figure 38, Western cooking emitted an average of 223.5 µg/m^{3}with concentrations ranging from 171.2-275.9 µg/m^3, Indian cooking emission average 183.2 µg/m^3 with a range of 148.9-241.1 µg/m^3, Chinese cooking 1009.5 µg/m^{3}with a range of 732.4 µg/m^3 to 1152.1 µg/m^3, African had average $PM_{2.5}$ concentration of 185.3 µg/m^3 and a range of 145.6 µg/m^3-268.1 µg/m^3.

It was therefore observed from these findings that Chinese cooking emits a higher concentration of $PM_{2.5}$ than the other food cooking styles evaluated. The Western style cooking emit less than the Chinese cooking and was observed to be the next higher concentration of $PM_{2.5}$ with similar concentration emitted during Indian and African style cooking. This showed a similar trend to what was observed in the trailer however it was found that the concentrations were about three fold more in the real kitchen (Chinese 1000µg/m^3 in kitchen and 370µg/m^3 at

cooking source). In the real kitchen there was no use of air extractor and no ventilation during sampling while in the trailer samples were collected in the duct of the extractor hood while the extractor was working. The idea behind taking samples in the duct and at a calculated distance was to obtain representative PM which had not undergone coagulation and condensation. In the real kitchen it could be assumed that due to the fact that samples were collected near the cooking point with all the heat and steam generated, more particles were collected in the general microenvironment. It was interesting to see that the trend was consistent for the cooking styles as was observed in the trailer in Table 20.

Similar to the finding in this research, a study by See and Balasubramanian (2006b) found that $PM_{2.5}$ mass concentration inside a food stall exceeded the 24 h standard by at least 400% during cooking hours suggesting that potential health risks exist in the Chinese food stall during cooking hours. The primary standards set are generally meant to protect public health with an adequate safety margin

Some studies have found low concentration of $PM_{2.5}$ for instance a study in the US found that the average $PM_{2.5}$ concentration due to cooking over 195 cooking events was about 5.5 µg/m3 with a standard error of 2.3 µg/m3 (Allen et al., 2004) , while in Europe, a study made involving the comparison of elderly residential homes in Amsterdam (47) and Helsinki (37), found that the estimated contribution from cooking ranged from 1.9 µg/m3 for indoor $PM_{2.5}$ in Helsinki to 3.4 µg/m3 for $PM_{2.5}$ personal exposure concentrations (Brunekreef et al., 2005).

Generally the rates of emission of aerosol have been reported to vary based on type of appliance used, the cooking conditions used and fat content of meat (McDonald et al., 2003). In an experiment where hamburger, steak and chicken were grilled and charbroiled, McDonald et al. (2003) found that the $PM_{2.5}$ emission rate for charbroiling meats ranged between 4.4 to 15 g/kg. The largest quantity of $PM_{2.5}$ was emitted by hamburger (15 g/kg) which had higher

fat content (30%) and were cooked on a char broiler. These results are consistent with data reported by Hildemann et al. (1991a). McDonald et al. (2003) reported that charbroiling produced higher concentrations than frying, 12-46 g/kg meat when charbroiling vs. 0.57 g/kg meat when frying. They also reported that charbroiling lean meat produced less concentrations of particles in the smaller size range (<20 nm) and in the larger size range (>100 nm) than regular meat. In this study it was found that stir frying in Chinese cooking produced higher concentrations of $PM_{2.5}$ than deep frying and stewing in Western and African and Indian style cooking.

Increased emissions measured at the source were reported to be a function of increased cooking temperature. Foods containing a higher percentage of fat generated higher emission rates than those with less fat percentage. They reported higher aerosol mass emission when cooking fatty foods (280-389 µg/m3) than when cooking vegetables (78 $\mu g/m^3$). Particle emission factor varied significantly also with type of oil used. Sunflower oil generated the lowest mass emission factors, whilst the highest emissions were from olive oil (Buonanno et al., 2009). Glytsos (2010) reported that frying of onions in olive oil in a controlled room emitted $PM_{2.5}$ in the range of 70 to 600 $\mu g/m^3$ (Glytsos et al., 2010).

See and Balasubramanian (2006b) investigated the physical and chemical properties of emissions from a Chinese food stall in Singapore while food was stir fried in a wok using a gas stove, and at two different and distinct times (See and Balasubramanian, 2006b). The mass concentration of particles ($PM_{2.5}$) measured in the food stall at the opposite site of a 4-LPG burner stove increased from 26.7 $\mu g/m^3$ during non-cooking hours to 312.4 $\mu g/m^3$ during cooking hours (increased by a factor of 12).

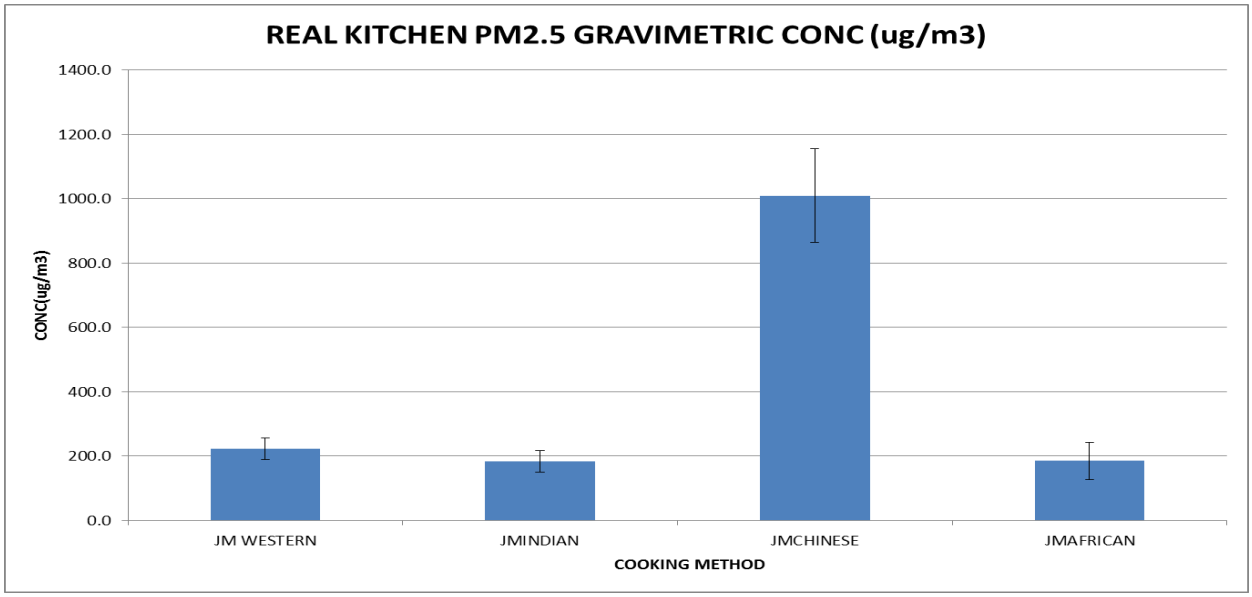
Analysis of various cooking methods which included steaming, boiling, stir-frying, pan-frying and deep-frying revealed that the largest amount of particulate matter measured at 20 cm from

the cooker was generated during deep frying ($PM_{2.5}$190 μg/m^3) and the lowest concentration was observed during steaming ($PM_{2.5}$72 μg/m^3) (See and Balasubramanian, 2008). Both studies have indicated that cooking with oil contributes to the production of more particles than cooking with water, which is consistent with the work of He et al. (2004a). In another study, See et al. (2006) made a comparison of emissions from Chinese, Indian and Malay food stalls and reported that the highest mass concentrations of $PM_{2.5}$ were found in the Malay stall (245.3 μg/m^3), whilst the lowest were measured in the Indian stall (186.9 μg/m^3) (See et al., 2006).

Similar to findings in this study several studies have found that Asian style cooking emits more particulate matter than Western cooking with concentrations of PM2.5 ranging 30 to 1,400 and 20 to 535 μg/m^3 as reported by various groups (Lee et al., 2001b; Levy et al., 2002; He et al., 2004b).

In California, a study was carried out in a test kitchen in a domestic setting where stationary samplers were positioned in the breathing zone of the cook to measure for UFP and $PM_{2.5}$ (Fortmann et al. 2001). It was found that the average PM concentrations when minced beef was pan fried with the ventilation system on and off were; 144 and 102 μg/m^3 ($PM_{2.5}$) and; 207 and 144 μg/m^3 (PM_{10}) respectively. Bacon was pan fried on a gas stove and was found to emit more $PM_{2.5}$ than on an electric stove; 482 μg/m^3 and 207 μg/m^3 respectively. Sampling was conducted in the nearby living room at the same time as cooking and higher concentration of $PM_{2.5}$ were observed when bacon was pan fried than when minced beef was fried, with 261 μg/m^3 and 7.8 μg/m^3 respectively (Fortmann et al. 2001, Sjaastad, 2010).

A summary of the studies discussed above is presented in Table 40.



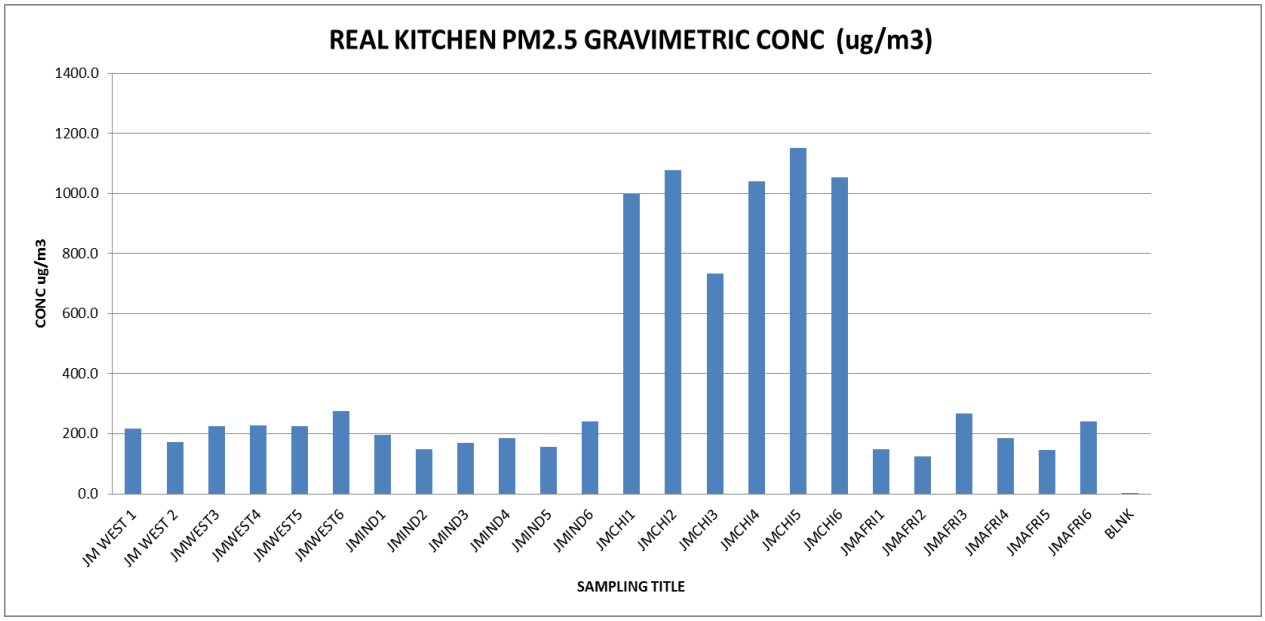


Figure 38 Gravimetric concentrations of $PM_{2.5}$ in real kitchen for 4 different cooking styles n=6 ($\mu g/m^3$)

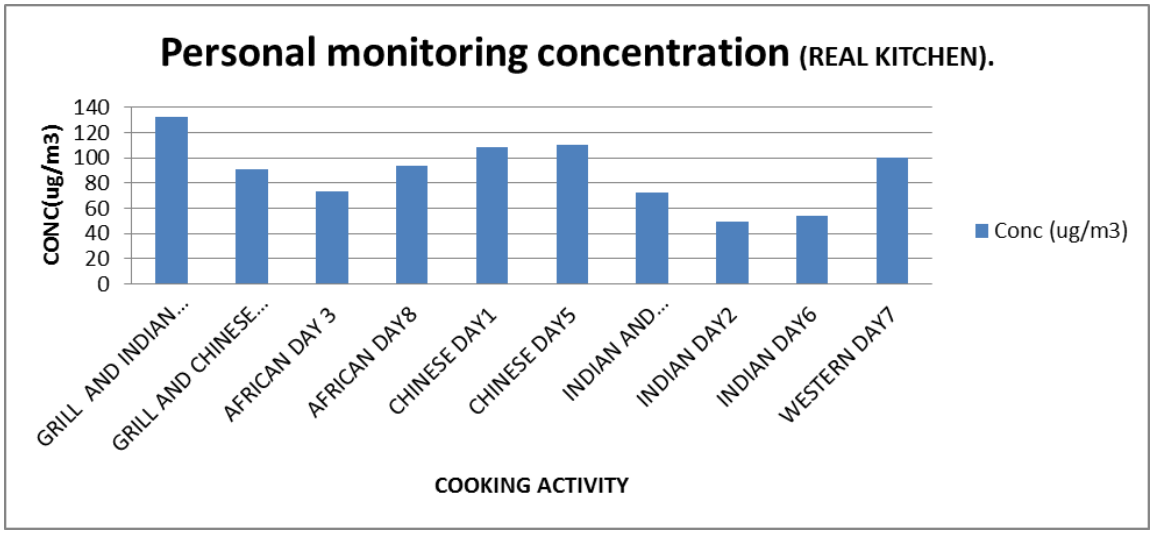


Figure 39 Gravimetric concentrations of PM from personal monitoring of cook in real kitchen (µg/m^3)

Figure 39 shows the concentration of PM obtained from personal monitoring of the cook for on a few of the days of sampling. The personal monitoring was carried out for 24 hours on days cooking activities took place. High concentrations are observed on the day that grilling was involved with a concentration of 120µg/m^3 on the day that grilling was carried out with Indian cooking. High PM concentrations were found on the das that Chinese cooking took place , consistent to all our findings of high loading of pm during Chinese cooking.

There is how ever an issue that as the sampler was worn for 24 hours there could be other factors that could affect the concentrations obtained as the cook goes about her daily chores. It is interesting to observe the trend is consistent with all the findings of the study.

Table 40 Concentration from previous studies

Reference	Location	Comment	Concentration (µg/m^3)
Abt et al. 2000	US	Frying - $PM_{0.02-0.5}$	29
		Frying – $PM_{0.7-10}$	19
		Barbequing - $PM_{0.02-0.5}$	57
		Barbequing - $PM_{0.7-10}$	12
		Oven cooking - $PM_{0.02-0.5}$	50
		Oven cooking – $PM_{0.7-10}$	8
		Sauteing - $PM_{0.02-0.5}$	42
		Sauteing – $PM_{0.7-10}$	294
		Toasting - $PM_{0.02-0.5}$	45
		Toasting – $PM_{0.7-10}$	8
Lee et al. 2001	China	$PM_{2.5}$ Chinese hot pot restaurant	81
		$PM_{2.5}$ Chinese dim sum restaurant	28.7
	Hong Kong	$PM_{2.5}$ Western Canteen	21.9
Levy et al. 2002	USA	$PM_{2.5}$ food court	200
Wallace et al., 2004	USA	Cooking dinner	
		Cooking breakfast	
He et al., 2004a	Australia	$PM_{2.5}$ (48h) cooking	37
		$PM_{2.5}$ (48h) cooking pizza	735
		$PM_{2.5}$ (48h) frying	745
		$PM_{2.5}$ (48h) grilling	718
		$PM_{2.5}$ (48h) kettle	13
		$PM_{2.5}$ (48h) microwave	16
		$PM_{2.5}$ (48h) oven	24
		$PM_{2.5}$ (48h) stove	57
		$PM_{2.5}$ (48h) toasting	35
		$PM_{2.5}$ residential kitchen	535.4
He et al., 2004c	China	$PM_{2.5}$ Hunan restaurant	1406
	China	$PM_{2.5}$ Cantonese restaurant	672
See and Balasubramanian, 2006a, See and Balasubramanian, 2008	Singapore	$PM_{2.5}$ Steaming	66 ± 7.6
		$PM_{2.5}$ Boiling	81 ± 9.3
		$PM_{2.5}$ Stir-Frying	120 ± 13
		$PM_{2.5}$ Pan-Frying	130 ± 15
		$PM_{2.5}$ Deep-Frying	190 ± 20
See and Balasubramanian, 2006b	Singapore	Stir-fry in a wok typical Chinese food commercial food stall $PM_{2.5}$	286
See et al., 2006	Singapore	$PM_{2.5}$ Chinese stall	202 ± 141
		$PM_{2.5}$ Malay stall	245 ± 77
		$PM_{2.5}$ Indian stall	187 ± 44
		$PM_{2.5}$ Background	29 ± 8
Buonanno et al., 2009	Italy	Grilling in a gas stove at maximum power	
		Cheese	283
		Wurstel sausage	352
		Bacon	389
		Eggplant	78

		Frying 50 g of chips in a gas stove at maximum power with Olive oil	118
		Peanut Oil	68
		Sunflower Oil	60
		Frying 50 g of chips using an electrical pan with sunflower oil	12
		olive Oil	27
		peanut Oil	13
Buonanno et al., 2010	Italy	PM_1 range	10-327
		$PM_{2.5}$	12-368
		PM_{10}	15-482
Glytsos et al. 2010	Czech Republic	Frying a slice of onion with olive oil – electric griddle	
Huboyo et al., 2011	Japan	Tofu boiling	22.8 (1.21-294)
		Tofu frying	41.2 (1.76-707)
		Chicken boiling	30.8 (5.36-1,082)
		Chicken frying	101.6 (1.67-1,366)
To and Yeung, 2011	Hong Kong	Frying vermicelli with beef – gas cooking (Domestic kitchen) – PM_{10}	1,330
		Frying vermicelli with beef – electric cooking (Domestic kitchen) – PM_{10}	1,030
		Pan Frying of meat – gas cooking (Domestic kitchen) – PM_{10}	1,020
		Pan Frying of meat – electric cooking (Domestic kitchen) – PM_{10}	520
		Deep frying of chicken wings – gas cooking (Domestic kitchen) – PM_{10}	890
		Deep frying of chicken wings – electric cooking (Domestic kitchen) – PM_{10}	680
		Deep frying of tofu – gas cooking (Commercial kitchen) – PM_{10}	4,720
		Deep frying of tofu – electric cooking (Commercial kitchen) – PM_{10}	3,980
		Griddle frying of meat – gas cooking (Commercial kitchen) – PM_{10}	2,260
		Griddle frying of meat – electric cooking (Commercial kitchen) – PM10	2,600

4.3.1 Concentration of organic compounds emitted from cooking in real kitchen.

Concentration of compounds when analysed are generally higher for Chinese cooking as observed for the $PM_{2.5}$ concentration as illustrated and shown Figure 40, Figure 41, Table 41 and Table 42.

PAHs-

The highest concentrations of PAH emitted were observed across all food cooking styles for dibenz(a,h)anthracene with the lowest concentration displayed by benzo(e)pyrene of 0.04 ng/m^3 with Indian cooking having very high concentration (0.69 ng/m^3) of this compound compared to other cooking styles.

Vainiotalo and Matveinen (1993) measured PAHs at the breathing zone of selected people working at five different kitchens where frying of meat was carried out. Sampling was conducted at 250-300^oC and for periods of 30 mins to 3 hours. Low concentration of PAH such as fluorene, phenanthrene, anthracene, pyrene, benzo[a]fluorene, chrysene, BaP and BghiP were detected in some samples in the range of 0.02-2.3 μg/m^3, and concentration of Naphthalene was found to be 1.6-25.6 μg/m^3 (Sjastaad, 2010).

See et al in 2006 also sampled cooks working in kitchens cooking Chinese, Malay and Indian food and found that the average particulate concentrations of naphthalene were 1.9, 2.8 and 3.9 ng/m^3 of respectively with BaP and acenaphthylene concentrations of 5.6, 16.0, 0.9 and 2.4, 5.6, 2.7 ng/m^3 respectively (See et al. 2006, Sjastaad, 2010). BaP concentrations were higher during Chinese cooking than for Indian cooking but in the present study its concentration were quite similar for all cooking methods with values of between 0.4- 0.6 ng/m^3.

Chen et al. (2007) compared the cooking emissions from Chinese, Western and Western fast food restaurants and reported that the total particulate phase PAH percentage in Chinese

cooking was the highest , in this study where Indian cooking is additionally analysed it was observed that the highest concentration of PAH was emitted during Indian cooking followed by Chinese style cooking. Zhu et al. (2009) indicated that particulate phase PAHs (PPAH) were predominantly absorbed on $PM_{2.5}$ with a 59-97% total particulate phase proportion. Particle size distribution analysis (Saito et al., 2014) showed that almost all PPAHs are concentrated on particles with diameters of <0.43 μm(Gao et al., 2015). Lu et al. (2011) reported that total concentration of 8 PAHs could range from 7.1 to 320 ng/m^3 and from 0.15 to 35 ng/m^3 in residential environments in China and Japan, respectively(Lu et al., 2011). The findings from this study are similar to what was observed in the Japanese residential environment but are generally slightly lower (0.25-2.06 ng/m^3).

Generally the lower concentration observed in the present study compared to some of the studies mentioned is due to the fact that they were carried out in commercial restaurants in most of the other studies.

Acids-

9-Octadecenoic acid was found to have the highest concentration of acid emitted during cooking with highest concentration released during western style cooking 5.62μg/m^3 (as observed in Table 42 and Figure 43). 9,12-Octadecadienoic acid is the next dominant acid that was measured with highest concentration observed during African style cooking with a concentration of 7.73 μg/m^3. In terms of acids concentration it was found to have higher concentrations measured across all the species during western style cooking (deep frying). All the cooking styles are found to have a similar concentration of hexadecanoic acid except African style.

Sterols-

Figure 42 and Table 42 show the range of sterols and glycerides measured during cooking in the kitchen. The glycerides emitted were in the range of 0.7-1.4 μg/m^3 with generally higher

concentration of 1-monoolein for all the cooking styles. Levoglucosan concentrations were within 1.1 to 2.3 µg/m^3 with the highest concentration observed during Chinese cooking probably owing to the higher amount of vegetables used in this cooking style. Similarly highest concentration of cholesterol (0.45 µg/m^3) was observed during Chinese style cooking.

Alkanes-

Figure 41 and Table 41 show the alkanes measured in the kitchen concentration. Nonacosane (C29) to tritriacontane (C33) were generally elevated across all cooking styles with a similar trend across all cooking styles for most compounds except for nonacosane and tritriacontane where chinese cooking was found to have high concentration (2.3ng/m^3 and 3.7 ng/m^3) while the other cooking styles had concentration of about 1ng/m^3 of nonacosane and 1.2 of tritriacontane

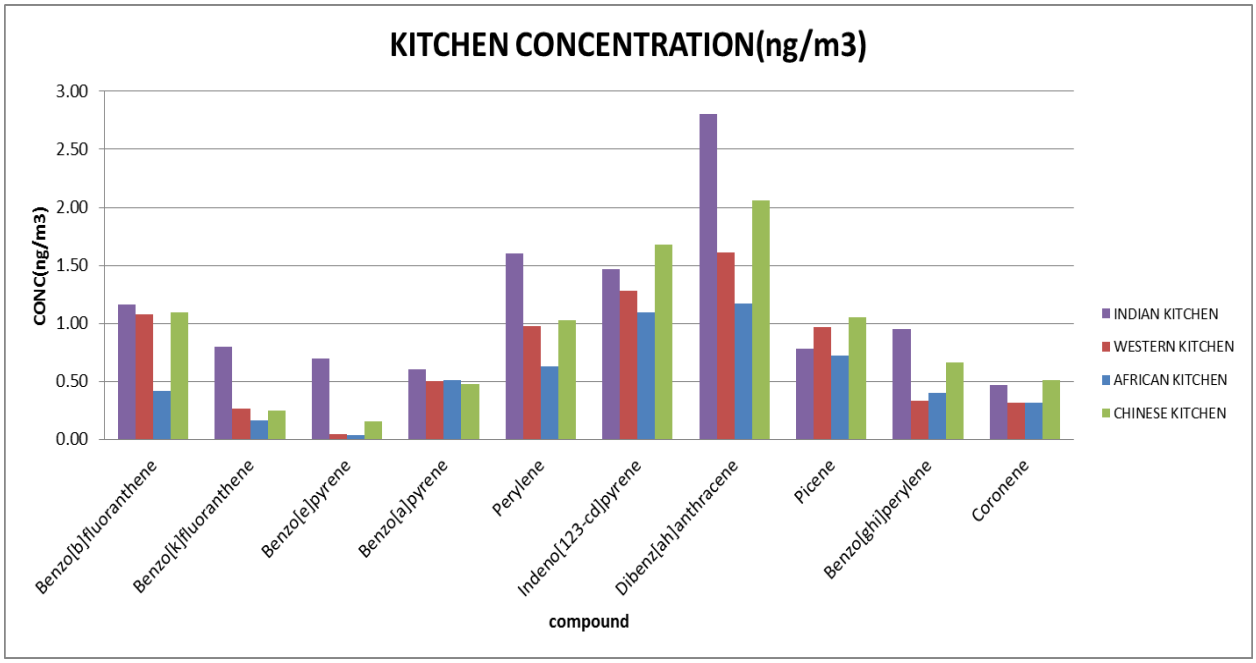


Figure 40 Concentration of compounds (PAH) emitted in real kitchen (ng/m^3)

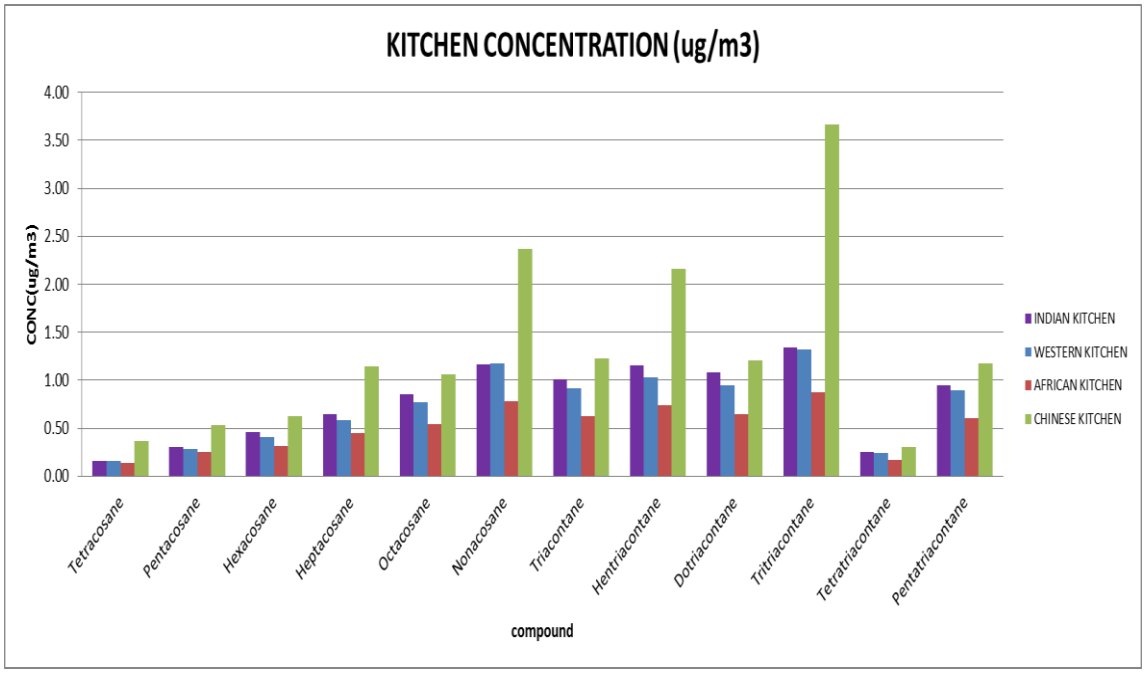


Figure 41 Concentration of compound (ALKANE) emitted in real kitchen (ng/m^3)

Table 41 Concentration of PAH and Alkane emitted in real kitchen (ng/m^3)

	INDIAN KITCHEN(n=6)							WESTERN KITCHEN(n=6)							AFRICAN KITCHEN(n=5)							CHINESE KITCHEN(n=6)						
PAH	average	std dev	MIN	25%	50%	75%	MAX	average	std dev	MIN	25%	50%	75%	MAX	average	std dev	MIN	25%	50%	75%	MAX	average	std dev	MIN	25%	50%	75%	MAX
Benzo[b]fluoranthene	1.16	0.92	0.45	0.62	0.45	1.57	2.74	1.07	0.56	0.34	0.60	0.34	1.45	1.58	0.42	0.23	0.21	0.29	0.35	0.44	0.79	1.09	0.43	0.57	0.75	0.57	1.35	1.68
Benzo[k]fluoranthene	0.80	1.36	0.07	0.09	0.07	0.68	3.51	0.26	0.26	0.07	0.07	0.07	0.47	0.62	0.16	0.11	0.07	0.07	0.15	0.19	0.33	0.25	0.21	0.07	0.07	0.07	0.39	0.56
Benzo[e]pyrene	0.69	1.05	0.01	0.04	0.01	0.96	2.62	0.04	0.06	0.01	0.01	0.01	0.01	0.15	0.04	0.02	0.01	0.01	0.05	0.05	0.05	0.15	0.13	0.01	0.06	0.01	0.21	0.35
Benzo[a]pyrene	0.60	0.44	0.31	0.38	0.31	0.54	1.48	0.50	0.28	0.20	0.27	0.20	0.72	0.85	0.51	0.35	0.11	0.21	0.54	0.78	0.93	0.48	0.16	0.24	0.39	0.24	0.60	0.67
Perylene	1.61	1.61	0.51	0.56	0.51	2.48	4.18	0.97	0.43	0.52	0.58	0.52	1.17	1.57	0.63	0.56	0.20	0.24	0.56	0.58	1.57	1.03	0.31	0.53	0.90	0.53	1.23	1.37
Indeno[123-cd]pyrene	1.47	1.10	0.69	0.83	0.69	1.58	3.58	1.28	0.49	0.41	1.40	0.41	1.52	1.61	1.10	0.74	0.25	0.67	0.86	1.69	2.03	1.68	0.46	1.08	1.44	1.08	1.88	2.30
Dibenz[ah]anthracene	2.80	2.81	1.07	1.33	1.07	2.61	8.39	1.61	0.51	0.85	1.54	0.85	1.87	2.25	1.17	0.60	0.45	0.74	1.14	1.74	1.80	2.06	0.56	1.25	1.75	1.25	2.33	2.84
Picene	0.78	0.26	0.48	0.58	0.48	0.90	1.18	0.97	0.50	0.42	0.56	0.42	1.32	1.60	0.72	0.29	0.43	0.45	0.72	0.90	1.10	1.05	0.39	0.50	0.93	0.50	1.11	1.70
Benzo[ghi]perylene	0.95	0.83	0.29	0.47	0.29	0.96	2.57	0.33	0.13	0.16	0.23	0.16	0.42	0.47	0.40	0.35	0.15	0.21	0.26	0.37	1.00	0.66	0.30	0.29	0.46	0.29	0.80	1.13
Coronene	0.47	0.52	0.12	0.16	0.12	0.46	1.37	0.32	0.29	0.01	0.11	0.01	0.52	0.63	0.32	0.29	0.00	0.09	0.28	0.53	0.69	0.51	0.24	0.18	0.40	0.18	0.63	0.88
ALKANES																												
Tetracosane	0.16	0.03	0.11	0.14	0.15	0.17	0.21	0.16	0.04	0.12	0.14	0.14	0.16	0.23	0.14	0.01	0.13	0.14	0.14	0.14	0.15	0.36	0.14	0.20	0.27	0.34	0.47	0.55
Pentacosane	0.31	0.06	0.25	0.27	0.28	0.35	0.39	0.28	0.06	0.24	0.25	0.27	0.30	0.39	0.25	0.02	0.23	0.25	0.26	0.26	0.28	0.53	0.13	0.36	0.44	0.53	0.61	0.70
Hexacosane	0.46	0.08	0.39	0.41	0.44	0.51	0.59	0.40	0.06	0.34	0.37	0.39	0.41	0.51	0.32	0.03	0.29	0.30	0.30	0.33	0.36	0.63	0.15	0.46	0.50	0.61	0.76	0.81
Heptacosane	0.65	0.12	0.56	0.56	0.58	0.72	0.83	0.58	0.08	0.49	0.54	0.57	0.59	0.72	0.45	0.05	0.37	0.44	0.47	0.48	0.50	1.14	0.22	0.83	1.04	1.10	1.31	1.40
Octacosane	0.86	0.14	0.75	0.76	0.78	0.96	1.04	0.77	0.10	0.69	0.73	0.75	0.77	0.96	0.54	0.06	0.43	0.54	0.55	0.57	0.61	1.06	0.22	0.79	0.92	1.02	1.21	1.36
Nonacosane	1.17	0.21	0.96	1.03	1.08	1.29	1.49	1.17	0.20	0.92	1.01	1.22	1.26	1.45	0.78	0.11	0.62	0.77	0.80	0.82	0.91	2.37	0.38	1.79	2.22	2.37	2.51	2.95
Triacotane	1.01	0.17	0.85	0.89	0.94	1.15	1.23	0.92	0.11	0.81	0.86	0.90	0.92	1.12	0.62	0.08	0.49	0.63	0.63	0.65	0.72	1.23	0.18	0.98	1.12	1.22	1.35	1.48
Hentriacotane	1.15	0.17	0.97	1.05	1.08	1.29	1.38	1.03	0.12	0.90	0.95	1.01	1.05	1.25	0.74	0.11	0.57	0.73	0.74	0.75	0.88	2.16	0.48	1.48	1.97	2.13	2.35	2.90
Dotriacotane	1.08	0.16	0.90	0.97	1.04	1.23	1.29	0.95	0.12	0.80	0.84	0.97	1.03	1.11	0.64	0.09	0.51	0.65	0.65	0.66	0.76	1.21	0.21	0.97	1.05	1.19	1.34	1.51
Tritriacotane	1.34	0.18	1.14	1.20	1.32	1.47	1.57	1.32	0.25	0.99	1.14	1.34	1.43	1.69	0.87	0.18	0.63	0.82	0.82	1.00	1.10	3.66	2.69	1.17	1.41	3.22	5.31	7.53
Tetratriacotane	0.25	0.13	0.00	0.26	0.29	0.33	0.35	0.24	0.03	0.21	0.22	0.24	0.24	0.30	0.17	0.02	0.13	0.16	0.17	0.17	0.20	0.30	0.04	0.26	0.27	0.30	0.31	0.38
Pentatriacotane	0.95	0.49	0.00	0.93	1.08	1.28	1.31	0.89	0.11	0.78	0.81	0.88	0.92	1.09	0.61	0.09	0.46	0.58	0.61	0.64	0.72	1.17	0.17	1.01	1.07	1.15	1.19	1.49

Table 42 Average concentrations of acids and sterols emitted in kitchen (μg/m^3)

μg/m^3	INDIAN KITCHEN(n=6)							WESTERN KITCHEN(n=6)							AFRICAN KITCHEN(n=5)							CHINESE KITCHEN(n=6)						
	average	std dev	MIN	0.25	0.5	0.75	MAX	average	std dev	MIN	0.25	0.5	0.75	MAX	average	std dev	MIN	0.25	0.5	0.75	MAX	average	std dev	MIN	0.25	0.5	0.75	MAX
Undecanoic	0.54	0.11	0.46	0.48	0.50	0.58	0.67	0.97	0.58	0.49	0.62	0.79	1.14	1.81	0.35	0.03	0.33	0.33	0.34	0.36	0.39	0.27	0.25	0.10	0.10	0.15	0.30	0.75
Octanedioic	0.32		0.32	0.32	0.32	0.32	0.32	2.43	2.08	0.07	1.67	3.27	3.62	3.96	0.04		0.04	0.04	0.04	0.04	0.04	0.25	0.18	0.09	0.13	0.16	0.37	0.52
Dodecanoic								1.83	1.72	0.54	0.86	1.18	2.48	3.78								0.14	0.10	0.00	0.06	0.17	0.19	0.27
Nonanedioic								0.44	0.22	0.28	0.36	0.44	0.51	0.59	0.23	0.04	0.20	0.20	0.20	0.20	0.25	0.10	0.08	0.00	0.05	0.10	0.12	0.23
Tridecanoic	0.19	0.24	0.06	0.06	0.06	0.26	0.46	0.45	0.72	0.03	0.09	0.12	0.49	1.53	0.22	0.21	0.08	0.37	0.37	0.37	0.37	0.44	#DIV/0!	0.44	0.44	0.44	0.44	0.44
tetradecanoic	0.13	0.03	0.11	0.12	0.12	0.14	0.17	1.14	1.85	0.04	0.19	0.20	0.87	4.41	0.17	0.04	0.14	0.14	0.17	0.18	0.20	0.19	0.07	0.11	0.16	0.17	0.21	0.31
pentadecanoic	0.43	0.18	0.10	0.42	0.49	0.54	0.57	2.11	2.32	0.16	0.69	1.03	3.10	6.06	0.23	0.10	0.06	0.25	0.25	0.26	0.31	0.61	0.16	0.40	0.52	0.61	0.70	0.82
Hexadecanoic	2.31	1.07	0.90	1.45	2.91	3.04	3.11	2.77	1.54	1.29	1.53	2.27	4.01	4.78	1.19	2.15	0.05	0.16	0.32	0.41	5.02	3.93	4.97	0.05	0.81	1.08	7.44	11.06
heptadecanoic	0.14	0.12	0.01	0.05	0.13	0.21	0.31	0.08	0.12	0.00	0.03	0.04	0.09	0.26	0.15	0.14	0.01	0.05	0.10	0.26	0.33	0.31	0.71	0.01	0.01	0.01	0.04	1.76
9,12-Octadecadienoic	0.57	0.57	0.12	0.20	0.42	0.62	1.65	5.62	10.74	0.20	0.27	0.63	3.79	27.25	3.30	4.74	0.02	0.05	0.73	4.68	11.04	4.04	7.70	0.06	0.42	0.84	2.12	19.65
9-Octadecenoic	2.62	2.14	0.02	0.97	2.83	3.81	5.57	2.93	3.17	0.23	0.92	1.24	5.28	7.41	7.73	1.82	6.27	6.70	7.02	7.81	10.82	5.11	5.08	1.15	1.64	2.83	7.40	13.68
Octadecanoic	0.38	0.42	0.01	0.04	0.29	0.56	1.08	2.82	4.38	0.10	0.39	0.93	3.37	9.34	0.92	0.31	0.51	0.83	0.92	0.99	1.37	2.21	1.71	0.01	0.78	2.88	3.31	3.98
nonadecanoic	1.61	2.35	0.00	0.08	0.15	3.39	4.83	0.06	0.06	0.01	0.02	0.04	0.09	0.16	0.16	0.11	0.01	0.09	0.18	0.22	0.28	0.26	0.43	0.03	0.07	0.11	0.13	1.15
Eicosanoic	0.77	0.70	0.10	0.15	0.69	1.40	1.50	0.40	0.56	0.05	0.09	0.22	0.32	1.51	0.87	1.28	0.06	0.15	0.34	0.69	3.13	1.26	1.49	0.22	0.33	0.38	2.02	3.71
Docosanoic	0.27	0.51	0.06	0.06	0.06	0.06	1.31	0.14	0.16	0.02	0.06	0.07	0.14	0.44	0.75	0.64	0.06	0.09	0.90	1.31	1.37	2.12	4.77	0.04	0.15	0.21	0.28	11.85
tetracosanoic	0.06	0.04	0.00	0.02	0.07	0.08	0.10	0.10	0.04	0.06	0.08	0.09	0.13	0.15	0.05	0.03	0.03	0.04	0.06	0.06	0.09	0.12	0.04	0.08	0.09	0.11	0.13	0.18
1-Monomyristin	1.00	0.62	0.55	0.58	0.70	1.26	2.06	0.82	0.32	0.52	0.62	0.73	0.91	1.41	1.03	0.92	0.32	0.46	0.71	1.09	2.59	1.20	0.28	0.81	1.13	1.18	1.22	1.70
1-Monopalmitin	1.36	1.16	0.65	0.71	0.89	1.28	3.66	1.12	1.02	0.54	0.69	0.70	0.84	3.18	0.68	0.49	0.39	0.42	0.48	0.56	1.54	1.09	0.37	0.66	0.77	1.12	1.33	1.57
1-Monoolein	1.14	0.73	0.56	0.81	0.85	1.12	2.58	1.47	0.46	1.08	1.21	1.33	1.52	2.35	1.23	0.50	0.48	0.49	1.05	1.43	1.52	1.10	0.35	0.71	0.79	1.12	1.31	1.58
1-Monostearin	1.10	0.62	0.66	0.71	0.89	1.11	2.30	1.28	0.60	0.82	0.84	0.96	1.78	2.05	1.39	0.51	0.42	0.56	0.89	1.30	1.63	0.81	0.16	0.64	0.66	0.81	0.95	0.99
Levoglucosan	1.10	0.62	0.66	0.71	0.89	1.11	2.30	1.70	0.43	1.36	1.44	1.51	1.80	2.49	1.19	0.42	0.79	0.96	1.02	1.31	1.86	2.28	0.45	1.70	1.97	2.24	2.66	2.80
Cholesterol	0.22	0.03	0.19	0.20	0.21	0.24	0.27	0.25	0.07	0.17	0.21	0.26	0.27	0.36	0.14	0.02	0.12	0.14	0.14	0.14	0.17	0.43	0.17	0.19	0.35	0.42	0.53	0.66

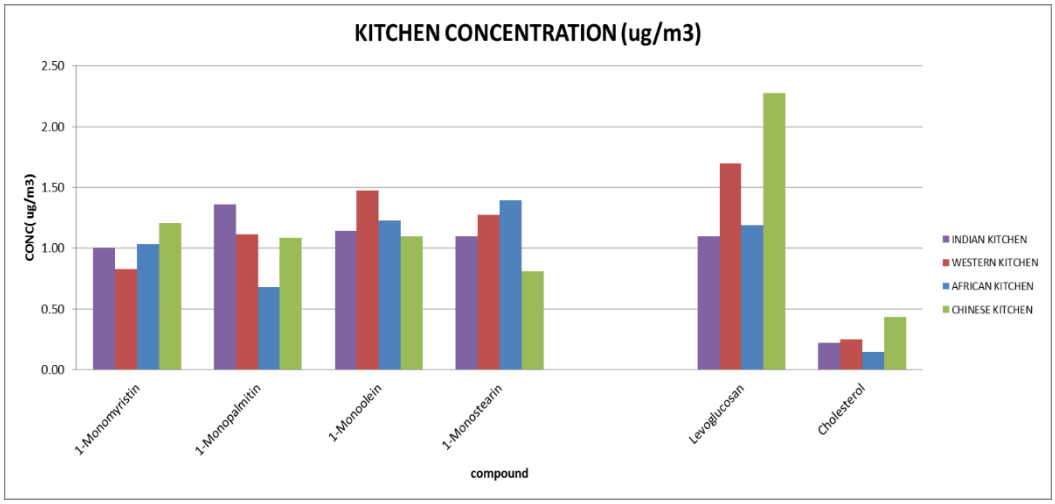


Figure 42 Concentration of sterol and glyceride emitted in real kitchen (µg/m^3)

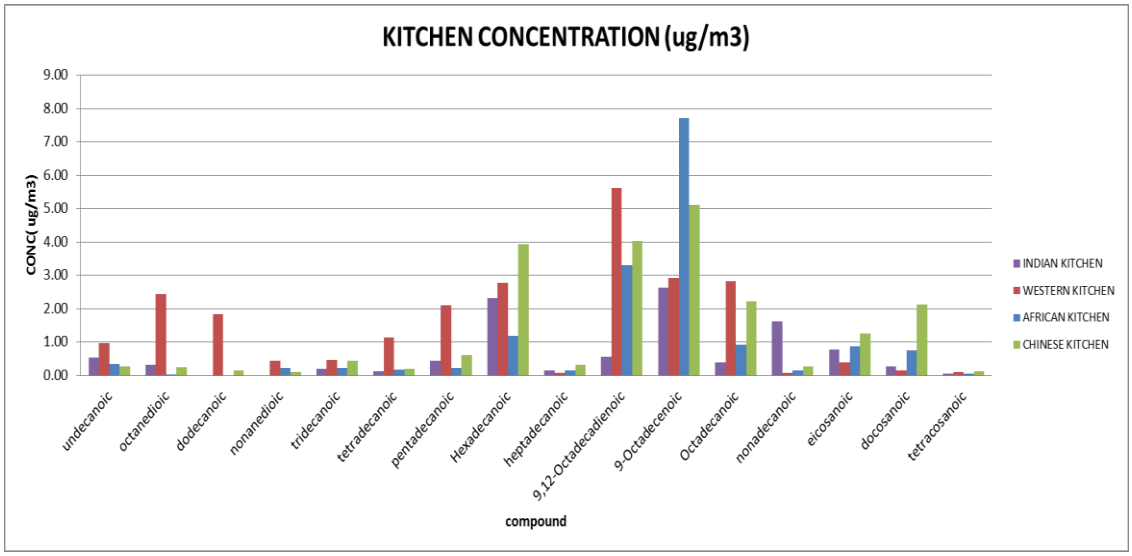


Figure 43 Concentration of acids emitted in real kitchen (µg/m^3)

Analysis of compounds emitted from the different cooking styles in Table 43, where a Spearman's rank correlation was run for the set of compounds against themselves across different cooking methods. Alkane compound concentration across all culinary techniques were well correlated with the highest correlation being between the African and western style as seen in Table 43A. Alkane compounds emitted from Chinese and Indian are significant but have the lowest correlation compared to the others with a value of 0.97.

In Table 43 B, acid concentrations were observed to show a different trend across the cooking methods with the highest correlation between African and Chinese cooking style with an r_s value of 0.7 ($p \leq 0.01$). The lowest correlation was found between Indian and Western cooking (r_s value of 0.41).

Rs value of 0.912 for African and Western cooking shows that the two cooking methods emissions were highly correlated with Chinese and Indian cooking in the kitchen found to also have good correlation for PAH with an r_s value of 0.8. African and Indian PAH concentrations were observed to have the least correlation across all styles of cooking with an r_s value of 0.69.

A correlation analysis of the cooking styles concentrations against the cooking profile concentrations obtained in chapter 3 are presented in Table 43(D), it was found that all the profiles are correlated with r_s values of between 0.52 -0.6 for all cooking styles against their respective profile concentration with African, Chinese, Western and Indian style having r_s values of 0.6, 0.52, 0.6 and 0.6 respectively.

These show that the profiles and concentrations are correlated at 99% significance related but with r_s value of 0.6 as such all have the chance of giving similar output when used in a CMB model.

Table 43 Correlation analysis of compounds emitted in kitchen

C. ALKANES

Correlations

		INDIAN	WESTERN	AFRICAN	CHINESE
Spearman's rho	INDIAN Correlation Coefficient	1.000	1.000**	1.000**	.979**
	Sig. (2-tailed)	.	.	.	.000
	N	12	12	12	12

		Correlation Coefficient	1.000**	1.000	1.000**	.979**
	WESTERN	Sig. (2-tailed)	.	.	.	.000
		N	12	12	12	12
		Correlation Coefficient	1.000**	1.000**	1.000	.979**
	AFRICAN	Sig. (2-tailed)	.	.	.	.000
		N	12	12	12	12
		Correlation Coefficient	.979**	.979**	.979**	1.000
	CHINESE	Sig. (2-tailed)	.000	.000	.000	.
		N	12	12	12	12

**. Correlation is significant at the 0.01 level (2-tailed).

D. ACIDS

Correlations

			INDIAN	WESTERN	AFRICAN	CHINESE
Spearman's rho	INDIAN	Correlation Coefficient	1.000	.407	.675**	.644*
		Sig. (2-tailed)	.	.149	.008	.013
		N	14	14	14	14
	WESTERN	Correlation Coefficient	.407	1.000	.625*	.529*
		Sig. (2-tailed)	.149	.	.013	.035
		N	14	16	15	16
	AFRICAN	Correlation Coefficient	.675**	.625*	1.000	.854**
		Sig. (2-tailed)	.008	.013	.	.000
		N	14	15	15	15
	CHINESE	Correlation Coefficient	.644*	.529*	.854**	1.000
		Sig. (2-tailed)	.013	.035	.000	.
		N	14	16	15	16

**. Correlation is significant at the 0.01 level (2-tailed).

*. Correlation is significant at the 0.05 level (2-tailed).

E. PAH

Correlations

			INDIAN	WESTERN	AFRICAN	CHINESE
Spearman's rho	INDIAN	Correlation Coefficient	1.000	.829**	.687*	.790**
		Sig. (2-tailed)	.	.003	.028	.007
		N	10	10	10	10
	WESTERN	Correlation Coefficient	.829**	1.000	.912**	.863**
		Sig. (2-tailed)	.003	.	.000	.001
		N	10	10	10	10
	AFRICAN	Correlation Coefficient	.687*	.912**	1.000	.867**
		Sig. (2-tailed)	.028	.000	.	.001
		N	10	10	10	10
	CHINESE	Correlation Coefficient	.790**	.863**	.867**	1.000
		Sig. (2-tailed)	.007	.001	.001	.
		N	10	10	10	10

**. Correlation is significant at the 0.01 level (2-tailed).

*. Correlation is significant at the 0.05 level (2-tailed).

F. ALL COMPOUNDS AGAINST SOURCE PROFILE CONCENTRATION

			AFRICAN	AFRIKIT	INDIAN	WESTERN	CHINESE	INDIANKIT	WESTKIT	CHIKIT
Spearman's rho	AFRICAN	Correlation Coefficient	1.000	.557**	.718**	.579**	.688**	.450**	.506**	.520**
		Sig. (2-tailed)	.	.000	.000	.000	.000	.003	.000	.000
		N	44	43	44	44	44	42	44	44
	AFRIKIT	Correlation Coefficient	.557**	1.000	.618**	.583**	.653**	.685**	.678**	.872**
		Sig. (2-tailed)	.000	.	.000	.000	.000	.000	.000	.000
		N	43	43	43	43	43	42	43	43
	INDIAN	Correlation Coefficient	.718**	.618**	1.000	.734**	.615**	.551**	.471**	.649**
		Sig. (2-tailed)	.000	.000	.	.000	.000	.000	.001	.000
		N	44	43	44	44	44	42	44	44
	WESTERN	Correlation Coefficient	.579**	.583**	.734**	1.000	.740**	.564**	.520**	.473**
		Sig. (2-tailed)	.000	.000	.000	.	.000	.000	.000	.001
		N	44	43	44	44	44	42	44	44
	CHINESE	Correlation Coefficient	.688**	.653**	.615**	.740**	1.000	.534**	.642**	.587**
		Sig. (2-tailed)	.000	.000	.000	.000	.	.000	.000	.000
		N	44	43	44	44	44	42	44	44
	INDIANKIT	Correlation Coefficient	.450**	.685**	.551**	.564**	.534**	1.000	.545**	.645**
		Sig. (2-tailed)	.003	.000	.000	.000	.000	.	.000	.000
		N	42	42	42	42	42	42	42	42
	WESTKIT	Correlation Coefficient	.506**	.678**	.471**	.520**	.642**	.545**	1.000	.525**
		Sig. (2-tailed)	.000	.000	.001	.000	.000	.000	.	.000
		N	44	43	44	44	44	42	44	44
	CHIKIT	Correlation Coefficient	.520**	.872**	.649**	.473**	.587**	.645**	.525**	1.000
		Sig. (2-tailed)	.000	.000	.000	.001	.000	.000	.000	.
		N	44	43	44	44	44	42	44	44

**. Correlation is significant at the 0.01 level (2-tailed).

The correlation shows of that the compounds and profiles are well correlated with r_s values of between 0.52 -0.6 for all cooking styles against their respective profile concentration with African, Chinese, Western and Indian style having r_s values of 0.6, 0.52, 0.6 and 0.6 respectively. These show that the profiles and concentrations are correlated at 99% significance related but with r_s value of 0.6 as such all have the chance of giving similar output when used in a CMB model.

Anova of the profiles showed that they are not significantly different with sig range-0.045-0.07.

4.4 Conclusion

Cooking was carried out in a real kitchen located in a regular house. The cooking was carried out without the use of an extractor fan and using gas as the energy source. Analysis of the filters that were collected after the sampling exercise higher gravimetric concentration during Chinese cooking consistent with findings in Chapter 3. Higher concentrations were found for 9-octadecenoic acid, levogluosan, tritricontane and dibenz{a,h}anthracene when the organic compounds collected on filters were analysed. This shows that these species could serve as good marker species for cooking. The analysis of samples collected from personal monitoring showed that the cook was exposed to higher concentration of PM on the days cooking of Chinese food was done. This further shows that Chinese cooking emits more organic matter than other cooking styles.

CHAPTER 5- Chemical mass balance model (CMB) modelling

This chapter contains some sections of verbatim text adapted from the following review article published as part of this PhD:

Abdullahi, L, Delgado Saborit, JM & Harrison, RM 2013, 'Emissions and indoor concentrations of particulate matter and its specific chemical components from cooking: A review' **Atmospheric Environment**, vol 71, pp. 260- 294.

5.1 Introduction

Information about certain sources of $PM_{2.5}$ have been found to be unavailable or weak such as solid fuel burning smoke (Harrison et al., 2012), cooking aerosol(Allan et al., 2010), abrasion particles from road vehicles (Thorpe and Harrison, 2008; Pant and Harrison, 2013) and secondary organic fractions(Yin et al., 2015).

For better understanding of various emission sources and their potential to emit ambient PM, the Air Quality expert group has recommended further analysis of these sources(AQEG, 2012). The information would be useful to the government and will be able to better forecast and thereby effectively plan and control emissions from these sources.

In previous studies, the source profiles generally used in CMB models include charbroiled meat cooking, gasoline vehicle emissions, diesel truck emissions and paved road dust (Hildemann et al., 1991a; Schauer et al., 1999a; Schauer et al., 1999b; Fraser et al., 2002) and for vegetative detritus and natural gas combustion (Rogge et al., 1993a; Rogge et al., 1993b). These profiles are mainly obtained from studies in the United States such as Texas and Los Angeles (Zheng et al., 2002; Fraser et al., 2003). However, the use of source profiles from locations other than the area of study might introduce uncertainty in the apportionment of some source contributions (e.g. road dust; soil).

A review by Lin et al., 2010 has also identified the need for more specified organic compound markers for some PM sources such as non-meat cooking particle emissions, paved roads,

fugitive dust, biogenic, and agriculture emissions as well as a source contributions library for particular locations, for use in CMB models.

Mass balance models that have been applied in indoor environments have usually taken into consideration various combustion related activities like home heating and cooking; and also activities such as cleaning and infiltration of outdoor air resulting in a contribution from outdoor sources (Millar et al., 2010). CMB analyses have made use of different combinations of source profiles for the estimation of the contribution of food cooking emissions to ambient particle concentrations. Several food cooking source profiles have been published (Rogge et al., 1991; Nolte et al., 1999; Schauer et al., 1999a; Rogge, 2000; Schauer and Cass, 2000; Chow et al., 2004; Robinson et al., 2006). These cooking profiles contain speciated organic data with a range of emission composition and rates mainly dependent on cooking technique and food type. The use of source profiles and fitting species require that the model must include all major sources and the species should be conserved during transport from source to receptor (Watson et al., 1998; Robinson et al., 2006). Organic molecular markers such as oleic acid, cholesterol and palmitic acids are used to estimate the contribution of food cooking emission to primary organic aerosol (Rogge et al., 1991; Schauer et al., 1999a; Robinson et al., 2006).

Using the chemical mass balance model to apportion for the sources of PM2.5 in a city has been exemplified by Schauer et al. (1996). They found that the organic carbon mass contribution of PM due to meat cooking was about 23% in Los Angeles, which was comparable to findings by Hildemann et al. (1991b) and Rogge et al. (1991) in earlier studies. The CMB approach was also used to find that meat cooking contributed between 20% and 75% to ambient concentrations of four ring PAHs measured in residential areas (Venkataraman and Friedlander, 1994).

Robinson et al. (2006) made use of the basic set of source classes and compounds developed by Schauer et al. (1996) and Schauer et al. (2000). The CMB analysis included source profiles

of eight source classes: diesel vehicles, gasoline vehicles, road dust, biomass combustion, cooking emissions, coke production, vegetative detritus and cigarettes. However, Robinson et al. (2006) reported that a large systematic bias was generally observed in CMB models due to differences in species and source profile marker to organic carbon ratios. The ambient ratio of palmitic acid to oleic acid was higher than expected from other published literature, reflecting problems presented by source profile variability. This signified that the CMB could not fit both the acids simultaneously, even though ambient concentrations showed a strong correlation indicating they were from the same source. The use of the two alkanoic acids as fitting species in the model in addition to other cooking markers however provided a better model for source contribution estimates, further highlighting the importance of molecular markers in source apportionment analysis. The model apportioned 320 ± 140 ng /m^3 (10% of the study average ambient organic carbon) to food cooking emissions.

Several other studies have illustrated the importance that cooking is a source contributing to organic aerosol (Ham and Kleeman, 2011, Wang et al., 2009, Hildemann et al.,1991b and Rogge et al.,1991). The studies have also identified that in order to reduce particle pollution, especially in populated metropolitan areas, efforts should focus on controlling cooking as well as other particle sources such as traffic emissions.

There has not been much analysis focused on the chemical characterization of PM from cooking in the UK with the only published receptor modelling studies using CMB using non-local source profiles(Yin et al., 2015, Yin et al., 2010).

Therefore with the source profiles developed in Chapter 3 and sampling data obtained from sampling at Stratford Road Birmingham, model runs are made here to analyse the effectiveness of the profiles to apportion the various pollution sources at the sampling location.

5.2 Description of CMB model

Source apportionment is defined as a method used to quantify the contribution that different airborne particulate matter sources make to their concentrations at receptor locations in the atmosphere (Kleeman, 2003). Source apportionment models attempt to re-construct the impacts of emissions sources based on ambient data registered at monitoring sites (Hopke and Song, 1997, Viana et al., 2008and Watson et al., 2002). There are three main methods of source apportionment which include the evaluation of monitoring data; receptor modelling which is based on the statistical evaluation of PM chemical data acquired at receptor sites and emission inventories or dispersion models (Viana et al., 2008).

The monitoring data technique of source apportionment involves the basic numerical analysis of measured data and evaluating how concentrations vary with time, meteorology, pollution contribution source (Lenschow et al., 2001, Escudero et al., 2007). The use of emission inventories and dispersion models involves the simulation of aerosol emission, formation, transport and deposition(Eldering and Cass, 1996). Receptor modelling on the other hand uses the principle that mass and species conservation can be assumed and a mass balance analysis can be used to characterise particulate air pollutant sources to quantify the contribution of each source to a particular pollutant (Hopke et al., 2006).

The chemical composition at receptor sites and source emissions are what are used as information in receptor models to understand the observed ambient concentrations and apportion the mass to different emission sources (Henry et al., 1984; Gordon, 1988; Hopke, 1991).

Traditionally there are two main receptor model techniques, Chemical Mass Balance (CMB) (Schauer et al., 1996; Watson, 1984) and multivariate statistical methods such as PMF (Hopke, 2003)(Viana et al., 2008). Other types of receptor models include Principal Component

Analysis (PCA), Multilinear Engine (ME), and UNMIX and hybrid models such as Constrained Physical Receptor Model (COPREM) (Watson et al., 2002; Viana et al., 2008).

The common principles for the receptor models are:

- assumption of constant source signature from the sources to the receptor,
- the optimization of linear combinations of different sources in order to minimize the difference between calculated values and experimental values.

Generally CMB is used when the sources are clearly defined and quantified and PMF and UNMIX are used when the sources are unknown (Clarke et al., 2012, Lee et al., 2008).

CMB uses measured fingerprints of source emissions (source profiles) to reconstruct atmospheric concentrations of chemical species (Friedlander,1973 and Yin et al., 2010). The input for the model consists of source profiles for various primary pollution sources and ambient measurement concentrations with their uncertainty; the final output provides approximations of contribution of each source identified at the measured location to the total mass measured (Pant et al., 2014).

The CMB model assumes that ambient concentrations are a linear sum of contributions of the known sources of pollution, as such source composition contributions for all contributing sources is essential to obtain relate the measured concentrations of compounds of the location (Pant and Harrison, 2012).

This model requires chemical source profiles which describe the specific chemical composition of emitted particles as input in order to quantify those sources within the data and apportion for the sources in the atmosphere. Source profile and fitting specie selection for CMB analysis is

a sensitive process which requires careful consideration as the profiles must be adequately different for all the sources included in the model to ensure proper apportionment at the receptor. The source emissions in the profile should not interact with each other during transport and also their chemical and physical properties should be practically constant during their transport from source to receptor (Chow and Watson, 2002). Some sources do not have existing source signatures or ones specific to the location being studied. In such cases profiles are borrowed from other cities with similar pollution sources, which may not represent the source of emissions in the sampling area of interest perfectly (Lee et al., 2008b; Pant and Harrison, 2012). Similarly when source profiles are too similar, the CMB model yields large uncertainties in source contributions (Chow and Watson, 2002). Generally the species used in most CMB models are from the source profiles available through the USEPA Speciate database. The source profiles consist of both organic and inorganic aerosol constituents (Schauer and Cass, 2000).

Table 44 Comparison between CMB and multivariate models (extracted from Pant, 2014)

CMB Model	Multivariate Models
i] A key prerequisite is detailed information about the sources/emission inventories as well as source profiles [ii] Only one sample is required [iii] Does not apportion the secondary aerosols [iv] Cannot take into account the time variation of the pollutant concentration or source emission [v] Only non-reactive, stable tracer species can be used [vi] Near collinearity among source profiles can result in negative source contributions	[i] Qualitative information about the potential sources is enough, useful for areas where detailed emission inventories are not available and source profiles are not required [ii] Require large numbers of samples [iii] Unable to account for spatial and temporal correlation between emissions (e.g. motor vehicle and road dust) or source identified may contain more than one source [iv] Often unable to produce a fine resolution of the sources [v] Some of the models allow negative contributions to sources which is physically impossible (e.g. PCA) [vi] Information like meteorological data, particle size etc can be incorporated in the analysis

Model output definitions (USEPA, 1997, Watson et al., 1998, Cooperenv, 2014)-

This section includes technical descriptions of the model output definitions, and hence, the text is copied verbatim from the authors Watson et al., 1998 and Cooperenv, 2014 as well as from the report on "Chemical mass balance receptor model version 8(CMB8)" published by the USEPA (1997).

Source Contribution Estimate- Contribution from the source type of the profile being used to the profile normalizing component.

Standard Error – "The uncertainty of the source contribution estimate (SCE), expressed as one standard deviation of the most probable SCE. The STD ERR is obtained from the uncertainty estimates of the receptor data and source profiles through the effective variance least-squares calculations. STD ERR is dependent on the uncertainties of the input data and the degree of similarity of the source profiles used for the model run".

CHI SQUARE is "the square root of the sum of the squares of the RATIO R/U that correspond to fitting species divided by the degree of freedom. The uncertainties of the calculated species concentrations affects its value. A large CHI SQUARE (>4.0) means that one or more of the calculated species concentrations differs from the measured concentrations by several uncertainty intervals".

"R-SQUARE Variance in ambient species concentrations explained by the calculated species concentrations. A low R SQUARE indicates that the selected source profiles have not accounted for the variance in the selected receptor concentrations".

Percent Mass is "the sum of SCE divided by the total mass or concentration. A value approaching 100% is desired however a value near 100% can be an indication that there is a problem as a poor fit can force a high %MASS".

5.3 Stratford Road concentration

Restaurants near sampling site:

Restaurants included a range of Italian, Chinese and Pakistani restaurants, with some- sandwich, pizza and kebab shop; - Indian Balti restaurant and curry houses; Steak house and some bakeries. Some of the restaurant distances from the sampling point are listed below in Table 45 and shown in the map in Figure 44. There is a general mix of different types or restaurants near the monitoring site ranging from Indian, Chinese, kebab houses, bakeries, pizza shops as well as sandwich shops. From **Table 45**, it was observed that Caspian pizza and Subway were the nearest restaurants to the sampling site (10 meters) but Subway was found to be just a sandwich shop with no proper cooking taking place in the store. In Caspian pizza on the other hand, frying of meat and chips and baking of pizza were among the cooking practices found to be taking place in store. The other restaurants nearest to the monitoring site from the map in Figure 44 (about 150 meters radius) were mainly Indian restaurants with; Osmani, Junaids kebab, Shahi Nan kebab being the nearest restaurants. The various steak houses and kebab shops (chicken hut(150 meters away), Yaqubs steak house(128meters) would have a lot of frying and grilling of meat and chips among other cooking methods during the time of operation of the restaurants.

From this analysis, it was observed that the predominant cooking style taking place in Stratford road area was the Indian and western style cooking (involving frying of chips, meat, chicken or fish).

An analysis of the PM samples collected at the location went further to confirm and show that these were the main cooking methods used by neighbouring restaurants.

Table 45 Restaurants and distance from sampling site at Stratford Road

NAME	TYPE	DISTANCE(meters)
Subway	SANDWICH	10
Caspian pizza	PIZZA AND KEBAB	10
Osmani	INDIAN	73
Junaid's Kebab House	INDIAN KEBAB	78
Shahi Nan Kebab	INDIAN	105
Babekhyber restaurant & takeaway	AFGHAN, INDIAN	115
Sher khan	INDIAN	115
The minar	INDIAN	115
Yaqub contemporary steak house	PIZZA AND STEAK HOUSE	128
Lahore Karachi Chinese	CHINESE AND PAKISTAN	138
Khan saab kitchen	INDIAN	148
Chicken hut	WESTERN	150
Peri peri original	INDIAN	150
Taste of Pakistan	INDIAN	300
Gateway to kyber	INDIAN	300
Rangin –persian restaurant	PERSIAN	300
Greggs bakery	BAKERY	300
Taste of Pakistan	INDIAN	300
Pasta di piazza la favourite	ITALIAN AND ASIAN	450
Al frash	INDIAN BALTI	450
Shahabs	INDIAN BALTI	450

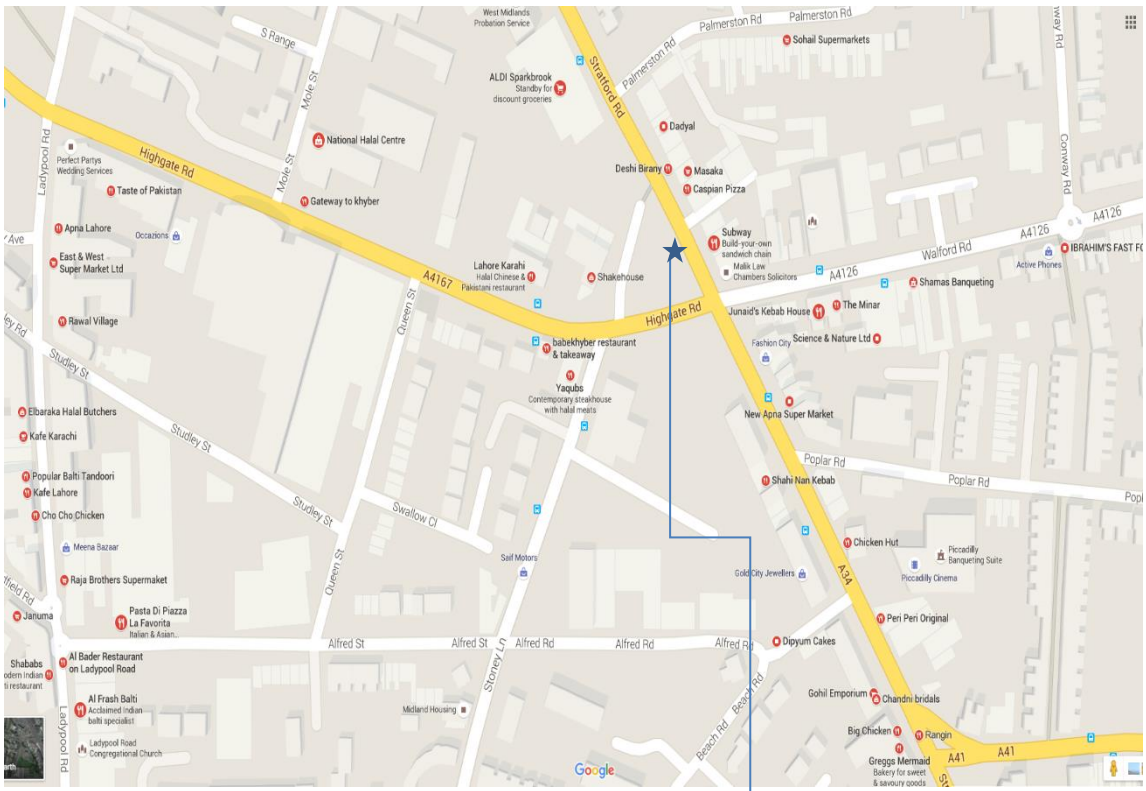


Figure 44 Map of Stratford Road showing restaurants and sample site. (Map data 2014: Google)

A dichotomous Partisol 2025 sampler and a Digitel DHA-80 sampler were collocated at the monitoring site for the purpose of gravimetric determination of the $PM_{2.5}$ mass concentration and organic analysis. The Stratford Road monitoring site is an urban roadside site, located on the kerb about 1meter of the road. The sampling interval was 24 hours daily from 12pm to 12pm between the 9th and 19th of December, 2014 and 9th and 19th of January, 2015. The Partisol sampler was used to collect samples onto 47 mm PTFE filters used for gravimetric and metal analyses while the Digitel was used for collecting also 24 h fine particles on 150 mm diameter quartz fibre filters, which were analysed for organic molecular markers, total organic carbon (OC), elemental carbon (EC),

The Partisol PTFE filters collected were conditioned and weighed in a controlled environment room (20 ± 2 ◦C and 35–45 % RH) before and after exposure to obtain the gravimetric mass of $PM_{2.5}$. After gravimetric analysis, those samples were analysed for elements Fe, Si and Al using a Bruker S8 Tiger WD-XRF (X-ray Fluorescence Spectrometer) instrument.

The Digitel $PM_{2.5}$ samples on quartz filters were analysed for OC and EC by Sunset Laboratory thermal-optical OC / EC analyser and organic markers by GC-MS.

From the sampling period, data from 13 days was used for the CMB model runs due to the availability of their subsequent element analysis data (the XRF Analysis was out sourced and was expensive so 13 days were choosen to represent the sampling campaign) an effort was made to represent and cover the sampling period .

5.4 Organic compounds

Concentrations of levoglucosan were between 9 to 65 ng m^{-3} with an average value of 22 ng m^{-3} for the sampling period at sampling site as shown in Table 46. It was observed that individual hopane concentrations were between 0.04 to 0.11 ng m^{-3} and PAH concentration were between 0.07 to 0.33 ng m^{-3}, these concentrations were similar to measured concentration

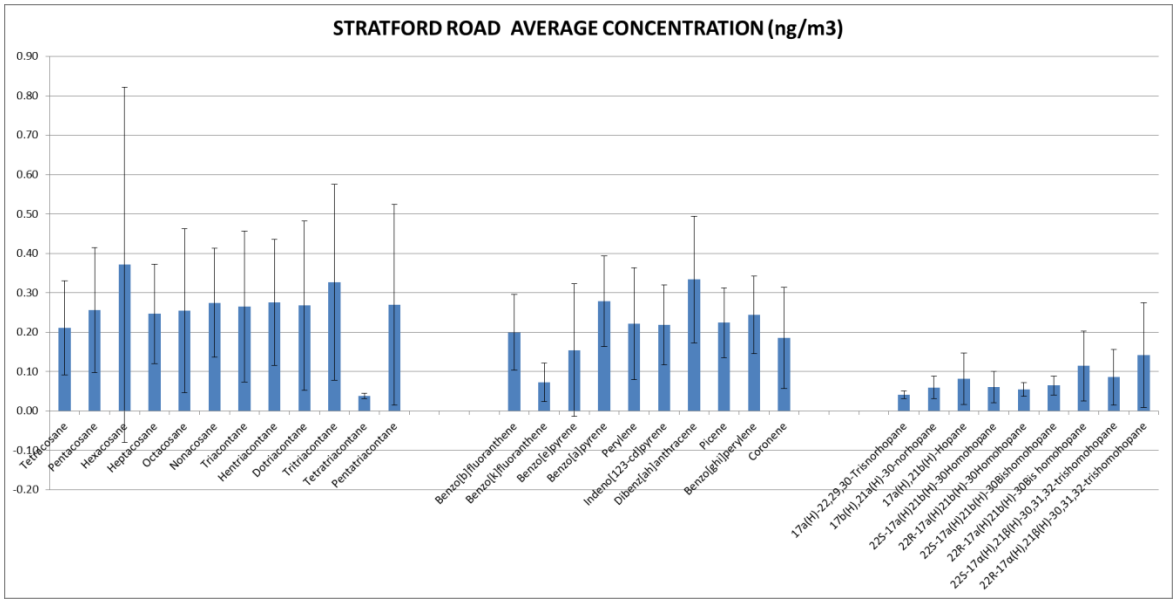
at the UK West Midlands urban background monitoring site, EROS, (levoglucosan 9.2 ng m^{-3} ; hopanes: 0.08– 0.18 ng m^{-3} ; PAHs: 0.06–0.27 ng m^{-3}) in 2007–2008(Yin et al., 2010). Higher levels were found in winter 2012 for levoglucosan (73.9 and 94.5 ng m^{-3}), hopanes (0.25–0.50 and 0.079–0.36 ng m^{-3}) and PAHs (0.10–0.67 and 0.044-0.51 ng m^{-3}) in Southeast England North Kensington sites and Harwell (Yin et al., 2015) than during the present sampling campaign at Stratford Road.

Alkane concentration in this study were between 0.23-0.37 ng m^{-3} which were low but similar to findings in 2012 in London (0.58–2.1 and 1.2–3.7 ng m^{-3} for NK and HAR). Alkane concentration were higher in winter periods in Birmingham 2008 at EROS (Elms road observatory site) (0.73–1.9 ng m^{-3}) and CPSS (Churchill pumping station site) (0.47–1.7 ng m^{-3}) (Harrison and Yin, 2010).

	09/12/2014	10/12/2014	11/12/2014	12/12/2014	13/12/2014	14/12/2014	15/12/2014	16/12/2014	17/12/2014	18/12/2014	09/01/2015	10/01/2015	11/01/2015	12/01/2015	13/01/2015	14/01/2015	15/01/2015	17/01/2015	18/01/2015
Tetracosane	0.85	0.96	0.80	0.41	0.90	0.56	1.00	0.74	0.47	0.39	0.48	0.34	0.46	0.72	0.73	0.50	1.41	1.26	1.75
Pentacosane	2.15	1.12	0.79	0.76	1.02	1.96	1.23	0.93	0.62	0.26	0.41	0.29	0.45	0.96	0.78	0.67	0.42	1.26	1.95
Hexacosane	1.23	0.53	1.61	0.85	1.04	2.99	1.12	0.77	0.43	0.90	1.28	0.53	8.69	0.83	0.74	0.59	1.09	0.63	1.80
Heptacosane	1.08	1.38	1.67	0.88	1.09	1.96	1.07	0.66	0.50	0.39	0.45	0.48	0.43	0.81	0.70	0.60	1.11	0.84	1.59
Octacosane	1.43	1.37	1.50	0.70	0.68	4.06	0.80	0.48	0.47	0.52	0.48	0.53	0.53	0.63	0.56	0.53	1.85	0.64	1.22
Nonacosane	1.90	1.44	1.93	0.83	0.91	2.48	0.95	0.63	0.65	0.63	0.66	0.95	0.63	0.80	0.73	0.74	1.21	0.81	1.07
Triacontane	2.40	1.09	2.25	0.72	0.61	3.22	0.67	0.59	0.58	0.60	0.61	0.81	0.67	0.62	0.61	0.59	1.99	0.60	0.84
Hentriacontane	2.63	1.40	2.02	0.64	0.76	2.17	0.86	0.65	0.63	0.64	0.71	0.68	0.61	0.75	0.70	0.70	1.93	0.75	0.99
Dotriacontane	2.34	1.49	2.20	0.77	0.58	3.81	0.65	0.58	0.59	0.63	0.63	0.65	0.59	0.61	0.57	0.59	1.90	0.63	0.69
Tritriacontane	3.39	2.36	2.82	0.75	0.73	3.65	0.86	0.69	0.70	0.76	0.80	0.92	0.68	0.72	0.67	0.68	2.33	0.73	0.75
Tetratriacontane	0.19	0.16	0.21	0.14	0.14	0.19	0.14	0.14	0.14	0.14	0.14	0.14	0.14	0.14	0.14	0.14	0.22	0.14	0.14
Pentatriacontane	3.50	1.55	1.11	0.67	0.59	3.74	0.58	0.59	0.55	0.65	0.59	0.75	0.58	0.55	0.53	0.51	2.82	0.55	0.58
Levoglucosan	5.72	18.80	6.20	55.81	12.03	12.88	10.25	8.91	9.06	33.03	65.02	6.77	21.26	17.24	36.13	37.62	12.72	12.87	50.02
Cholesterol	1.29	1.11	1.32	1.31	1.47	1.49	1.24	1.32	1.15	1.14	1.12	1.16	1.13	1.20	1.14	1.19	1.42	1.17	1.29
17a(H)-22,29,30-Trisnorhopane	0.14	0.14	0.23	0.18	0.18	0.13	0.17	0.15	0.12	0.13	0.14	0.13	0.13	0.17	0.14	0.13	0.26	0.16	0.22
17b(H),21a(H)-30-norhopane	0.15	0.16	0.24	0.29	0.22	0.16	0.29	0.19	0.16	0.16	0.16	0.13	0.14	0.25	0.20	0.22	0.45	0.23	0.40
17a(H),21b(H)-Hopane	0.28	0.21	0.42	0.37	0.30	0.20	0.26	0.20	0.18	0.14	0.17	0.16	0.17	0.32	0.27	0.25	1.33	0.23	0.44
Picene	1.50	1.08	0.96	0.98	0.93	0.69	0.93	1.25	0.42	0.97	0.89	0.75	0.54	0.52	1.03	0.00	1.04	1.27	1.42
Benzo[b]fluoranthene	0.71	0.82	0.55	0.89	1.49	0.26	1.24	1.33	0.41	1.01	0.95	0.60	1.29	0.39	0.97	0.00	0.85	0.96	0.77
Benzo[k]fluoranthene	0.47	0.08	0.05	0.19	0.57	0.27	0.09	0.21	0.40	0.57	0.34	0.20	0.38	0.48	0.14	0.00	0.23	0.65	0.07
Benzo[e]pyrene	0.46	1.20	0.00	0.45	0.44	0.03	1.97	0.86	0.37	0.34	0.04	0.02	0.46	1.43	0.12	0.00	0.03	0.66	2.30
Indeno[123-cd]pyrene	1.06	0.87	0.64	0.97	1.21	0.81	0.85	0.82	0.88	1.36	0.96	0.84	0.30	0.90	0.37	0.00	1.21	1.95	0.89
Benzo[ghi]perylene	0.78	0.67	0.83	1.45	1.31	1.12	1.02	1.06	1.02	1.02	1.65	1.02	0.87	0.58	0.66	0.00	1.39	1.59	0.53
Hexadecanoic	19.63		1.92	12.45	20.43	12.68	23.91		35.03	8.15		27.88	13.55	52.99	9.70	7.98	5.11		18.40
9,12-Octadecadienoic	0.58	0.30	0.11	0.22	0.14	0.20	0.51		0.83	0.35	0.20	0.19	0.28	3.65	0.33	0.31	0.15	0.07	0.54
9-Octadecenoic	5.31				6.08						0.00	18.17			0.16	5.26	2.82		
Octadecanoic	10.67		1.43	6.50	7.90	7.45	19.23		19.29	9.38		24.15	1.42		0.85	6.85	4.07		12.73
docosanoic	0.50	0.72	0.29	0.76	0.87	0.52	1.04	0.51	1.14	1.01	0.48	0.73	0.97	1.39	1.52	0.76	0.40	0.97	1.30
tetracosanoic	0.40	1.34	0.57	1.16	1.33	1.15	1.74	1.29	2.17	1.50	1.22	1.47	1.67	1.81	2.23	1.66	0.52	1.20	1.79
1-Monomyristin	17.09	8.03	3.43	2.88	7.52	9.58	5.11	33.25	6.67	4.22	4.31	11.58	3.17	7.04	2.90	9.26	9.46	7.64	2.91

1-Monopalmitin	19.00	4.63	3.51	2.77	6.20	20.79	6.01	10.40	14.13	15.18	2.99	22.40	3.15	9.86	2.76	7.45	26.46	8.23	2.79
1-Monoolein	18.78	3.17	5.43	3.13	4.92	28.39	7.55	67.47	12.69	9.10	3.23	24.02	3.33	6.54	3.13	8.01	14.18	21.31	3.16
1-Monostearin	34.26	3.61	5.61	2.82	3.76	16.50	6.57	15.87	28.28	8.22	2.95	31.78	3.01	7.00	2.82	6.39	6.46	10.18	2.85

Table 46 Daily concentration Statford road ng m^{-3}



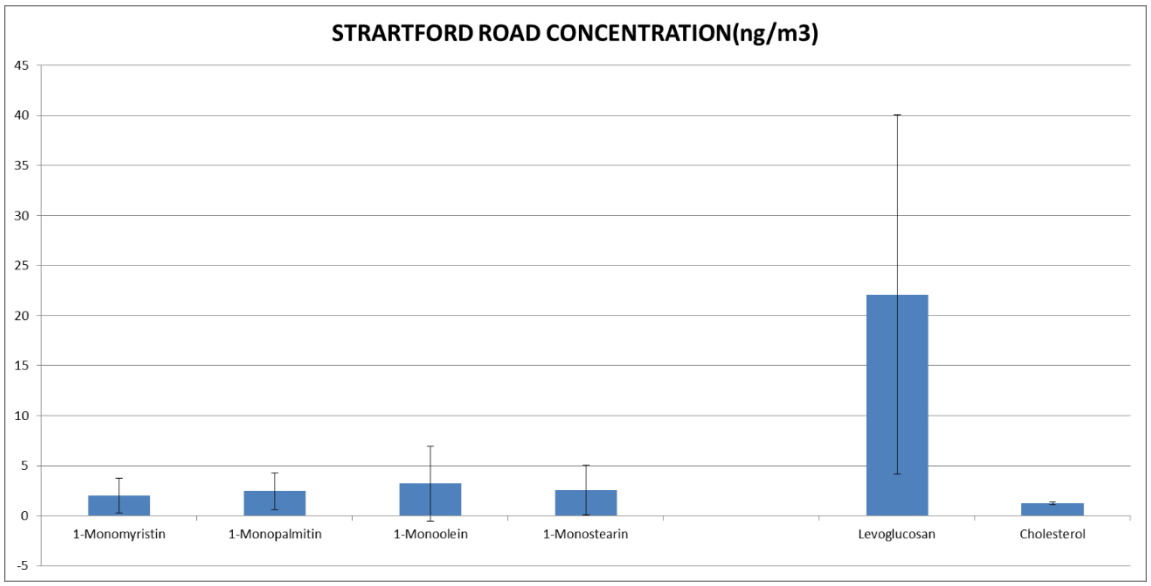


Figure 45 Stratford Road concentrations

A significant fraction of $PM_{2.5}$ atmospheric particles is characterized by carbonaceous content, about 21-78% of their mass (Na et al., 2004, Querol et al., 2004, Yu et al., 2004).

Elemental carbon (EC) is produced from the burning of carbonaceous matter while Organic carbon (OC) may be emitted directly in the particulate phase or formed reactions in the

atmosphere (Jones and Harrison, 2005; Saylor et al., 2006). EC has been found to be a good indicator of urban emissions from road transport (Gelencsér et al., 2007)

Table 47 represents the data for the OC/EC analysis as well as the gravimetric $PM_{2.5}$ concentration for the sampling days at Stratford Road. It was observed that on some days the $PM_{2.5}$ data obtained were unduly low and the error sources are not known for instance on 15/01/2015. The data for these days have still been analysed in the CMB model and have no implication on the general model run as each day is analysed independently.

Table 47 OC/EC and PM2.5

	OC(ug/m^3)	EC(ug/m^3)	total	PM2.5 (gravimetric)	EC/OC
10/12/2014	1.6	0.9	2.5	6.6	0.6
11/12/2014	1.6	1.1	2.7	9.0	0.6
13/12/2014	3.4	1.6	5.0	7.6	0.5
14/12/2014	1.4	0.9	2.3	7.0	0.6
15/12/2014	2.9	2.0	4.9	7.5	0.7
16/12/2014	1.7	1.1	2.8	8.9	0.7
17/12/2014	1.7	1.2	2.9	7.0	0.7
18/12/2014	1.4	0.8	2.2	5.3	0.6
09/01/2015	1.2	0.4	1.6	3.5	0.3
13/01/2015	3.6	1.6	5.3	4.7	0.4
14/01/2015	2.3	1.2	3.4	7.9	0.5
15/01/2015	4.7	1.3	6.0	7.0	0.3
18/01/2015	3.6	2.4	5.9	8.0	0.7

Table 48 Source profiles

	WEST	ARF	CHI	IND		WEST	ARF	CHI	IND
EC	2.66E-03	1.21E-03	3.19E-03	3.79E-03	DODE	7.49E-03	3.06E-03	5.46E-03	9.45E-03
ECU	1.00E-08	1.00E-08	1.00E-08	1.00E-08	DODEU	5.61E-03	2.51E-03	6.83E-03	3.18E-03
LEVOG	1.17E-02	6.14E-03	5.25E-03	1.75E-02	NONDIA	2.28E-03	1.01E-02	2.18E-03	2.03E-03
LEVOGU	6.26E-03	4.08E-03	2.67E-03	3.62E-03	NONDIAU	1.25E-03	1.33E-02	2.03E-03	1.96E-03
CHOL	2.30E-03	1.17E-03	8.02E-04	2.47E-03	TRI	6.10E-03	3.65E-04	1.27E-04	1.53E-03
CHOLU	3.18E-04	4.09E-02	2.37E-02	5.90E-02	TRIU	9.54E-03	3.12E-04	1.14E-04	8.46E-04
PICENE	2.56E-03	1.09E-03	1.29E-03	2.27E-03	TETDE	9.69E-03	9.43E-04	1.61E-03	3.32E-03
PICENEU	1.61E-03	1.26E-03	4.18E-04	2.72E-03	TETDEU	1.46E-02	1.16E-03	1.72E-03	2.43E-03
BZBFLU	2.17E-03	6.29E-04	5.68E-04	8.90E-04	PENT	6.07E-03	2.81E-03	1.81E-03	7.90E-03
BZBFLUU	1.46E-03	5.79E-04	2.62E-04	7.93E-04	PENTU	4.03E-03	2.40E-03	1.07E-03	3.71E-03
BZKFLU	1.57E-04	2.48E-04	2.15E-04	5.25E-04	HEP	5.57E-04	2.32E-04	1.45E-04	1.85E-04
BZKFLUU	6.82E-05	1.92E-04	3.08E-04	4.91E-04	HEPU	7.17E-04	1.89E-04	6.97E-05	6.10E-05
BZEPYR	1.58E-04	4.02E-04	4.55E-04	1.43E-04	NONA	2.49E-04	1.75E-04	1.79E-04	2.88E-04
BZEPYRU	3.08E-04	5.97E-04	4.65E-04	2.47E-04	NONAU	4.59E-05	1.06E-04	1.51E-04	2.29E-04
INDPYR	1.98E-03	5.87E-04	5.68E-04	1.12E-03	EICO	2.09E-04	2.94E-04	4.13E-04	3.75E-04
INDPYRU	1.49E-03	3.82E-04	1.91E-04	8.74E-04	EICOU	1.92E-04	3.59E-04	2.96E-04	1.85E-04
BZGHPL	1.68E-03	4.95E-04	1.05E-03	1.24E-03	DOCO	6.01E-03	5.13E-04	4.70E-04	5.07E-03
BZGHPLU	1.22E-03	3.58E-04	5.30E-04	1.04E-03	DOCOU	6.94E-03	5.40E-04	1.49E-04	5.53E-03
PALMTA	1.70E-02	2.59E-02	1.40E-02	1.99E-02	TETCO	1.04E-03	4.10E-04	3.96E-04	8.35E-04
PALMTAU	1.88E-02	3.77E-02	2.33E-02	3.92E-02	TETCOU	4.30E-04	2.19E-04	3.73E-05	2.56E-04
LINOLA	1.32E-02	1.12E-02	2.75E-02	1.77E-02	MONMY	3.71E-02	6.10E-03	1.23E-02	1.59E-02
LINOLAU	1.53E-02	1.47E-02	4.87E-02	2.35E-02	MONMYU	2.21E-02	5.79E-03	1.29E-02	9.00E-03
OLA	2.75E-02	2.28E-02	2.27E-02	4.14E-02	MONPA	4.46E-02	4.71E-03	2.79E-02	1.24E-02
OLAU	2.44E-02	3.03E-02	2.61E-02	4.03E-02	MONPAU	4.60E-02	3.69E-03	4.40E-02	7.02E-03
STEARA	2.87E-03	9.70E-03	8.64E-03	6.91E-03	MONOL	4.42E-02	1.21E-02	2.42E-02	1.48E-02
STEARAU	3.08E-03	7.35E-03	9.78E-03	7.57E-03	MONOLU	3.64E-02	1.38E-02	3.72E-02	1.07E-02
UNDEC	2.30E-02	1.71E-03	1.35E-03	4.70E-03	MONSTE	2.31E-02	9.50E-03	1.51E-02	1.58E-02
UNDECU	2.56E-02	1.26E-03	6.75E-04	5.17E-03	MONSTEU	5.10E-03	9.33E-03	1.71E-02	8.97E-03
OCTA	1.28E-02	3.49E-03	5.37E-03	2.12E-03					
OCTAU	1.20E-02	5.52E-03	7.42E-03	1.39E-03					

5.5 Model results

The CMB 8.2 model from USEPA was used for the estimation of source contribution to $PM_{2.5}$. The source profiles included in the model were; vegetative detritus (Rogge et al., 1993a), natural gas combustion (Rogge et al., 1993b), wood smoke/biomass burning (Fine et al., 2004; Sheesley et al., 2007), dust/soil (Sheesley et al., 2007), coal combustion (Zhang et al., 2008). A single traffic source profile was used for the model run, which was generated from a twin site measurement from London by Pant et al.,(2014) as it provided a better representation of the UK fleet and the older source profiles tended to overestimate the emissions from traffic (Yin et al., 2010; Pant et al., 2014). The source profile obtained from this study has been used

for the CMB modelling in order to obtain a better estimate and apportionment of the cooking profiles. As four different profiles have been obtained which represent different cooking styles and methods, the idea was to see how the model output would differ for the different cooking profiles.

The fitting species used in the model for this study included elemental carbon, silicon, aluminium, levoglucosan, C25-C35 alkanes, 17a(H)- 22,29,30-trinorhopane, 17a(H)-21b(H)-hopane, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[e]pyrene, indeno(1,2,3-cd) pyrene, benzo(ghi)perylene, picene, n-hexadecanoic acid, n-octadecanoic acid, 9-octadecenoic acid, 9,12-octadecadienoic acid, 1-Monopalmitin,1-Monostearin, 1-Monomyristin and 1-Monoolein .

The model run output was evaluated using certain parameters which include, goodness- of fit parameters- r2 and chi2 with acceptance of an r2 value between 0.8-1.0 and a chi-square value less than 4. Profiles with a negative source contribution were removed from subsequent runs, also the ratio of the source contribution estimate and standard error (t_{stat}) a value below 1 indicates the source is not significant and is below detection limit Species selected for use in the model included those with calculated and measured concentration ratio(C/M) between 0.75-1.5, species with ratio of signed difference between calculated and measured concentration R/U ratio between -2- +2. The markers for the different sources were monitored to identify the influential species for each source type and cross-validated with published marker data using the MPIN matrix (modified pseudo inverse normalized) matrix in the CMB model runs output. The influential species have values between 0.5 to 1in the MPIN matrix.(USEPA, 1997).

Table 49 Key markers used for the sources (based on MPIN matrix)

Source	Key Marker (value of 1.00)
Wood Smoke	Levoglucosan
Road dust	Si
Traffic	Benzo(ghi)perylene/ EC/Hopanes
Cooking	Cholesterol

Table 50 Source contribution estimate for average concentration at Stratford road (µg/m^3).

A. SCE $PM_{2.5}$

	SCE $PM_{2.5}$			
	Woodsmoke	DIRT/soil	TRAFFIC	COOK
WEST	0.17	0.15	0.55	0.10
INDIAN	0.16	0.15	0.55	0.10
CHINESE	0.17	0.15	0.55	0.10
AFRICAN	0.17	0.15	0.55	0.10

B. SCE OC

	SCE OC			
	Woodsmoke	DIRT/soil	TRAFFIC	COOK
WEST	0.14	0.20	0.33	0.13
INDIAN	0.14	0.20	0.33	0.12
CHINESE	0.15	0.20	0.33	0.13
AFRICAN	0.14	0.20	0.33	0.12

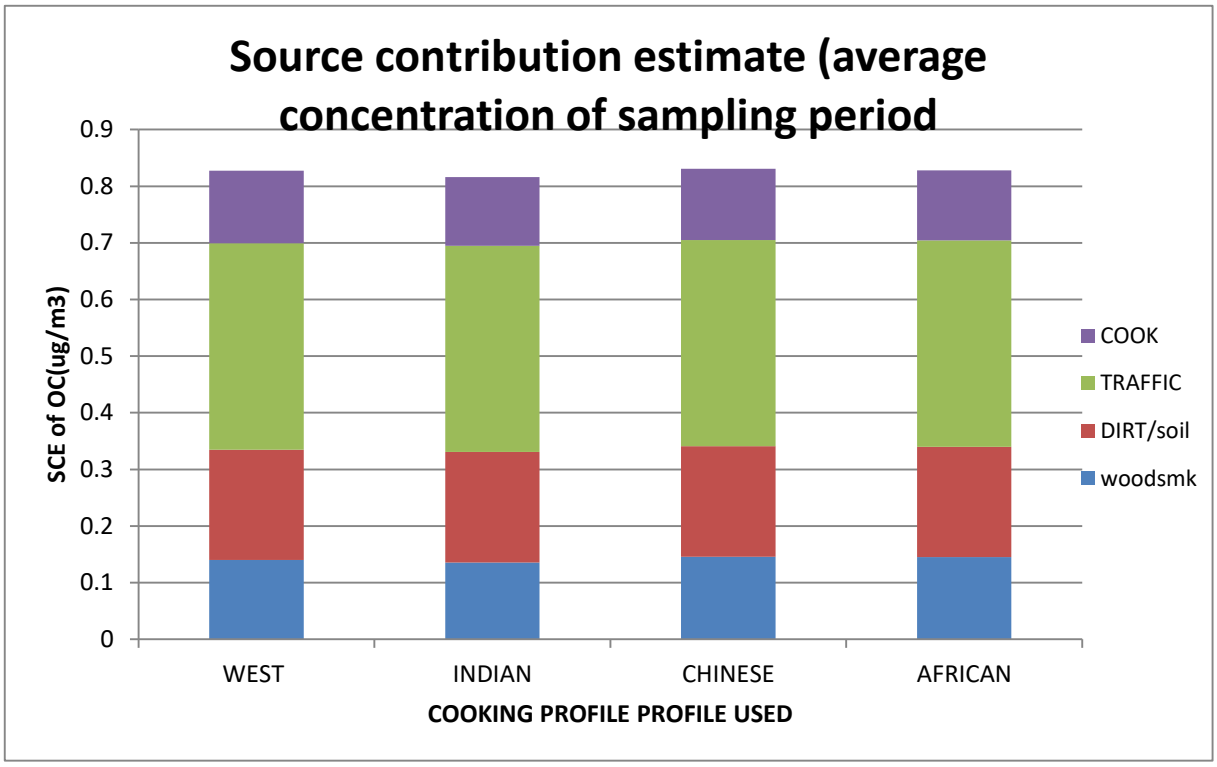


Figure 46 Average source contribution for average sampling period at Stratford road

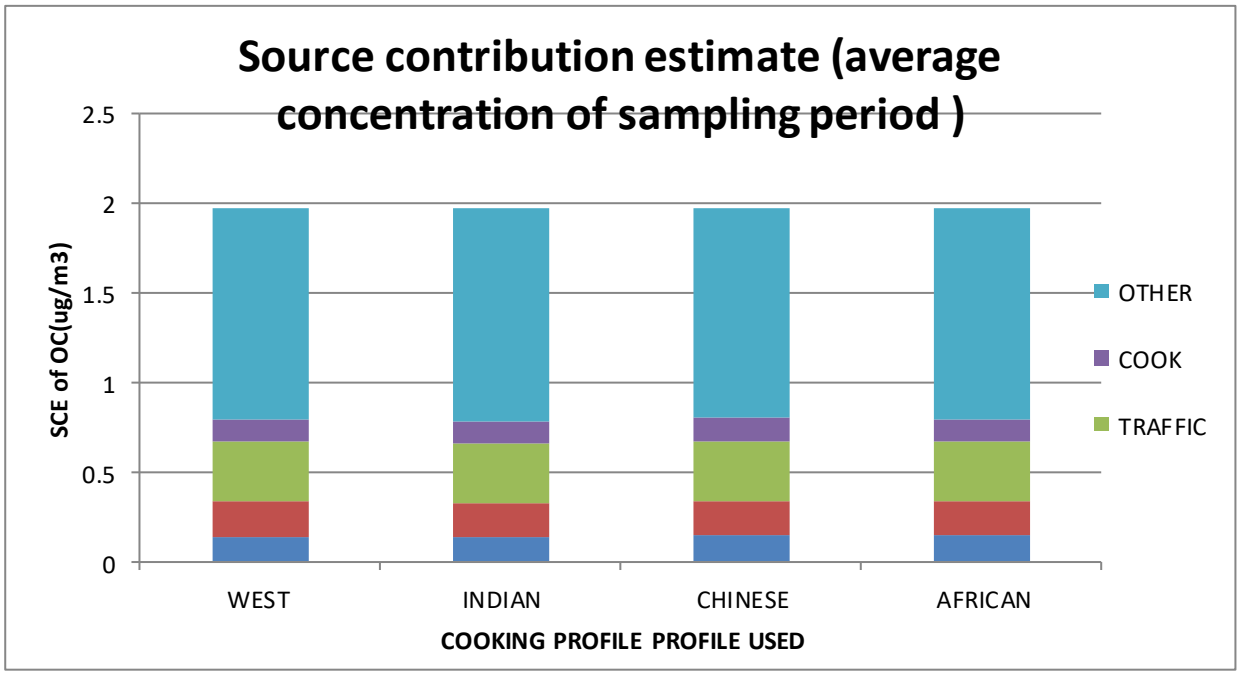


Figure 47 Average source contribution for average sampling period at Stratford road with other sources

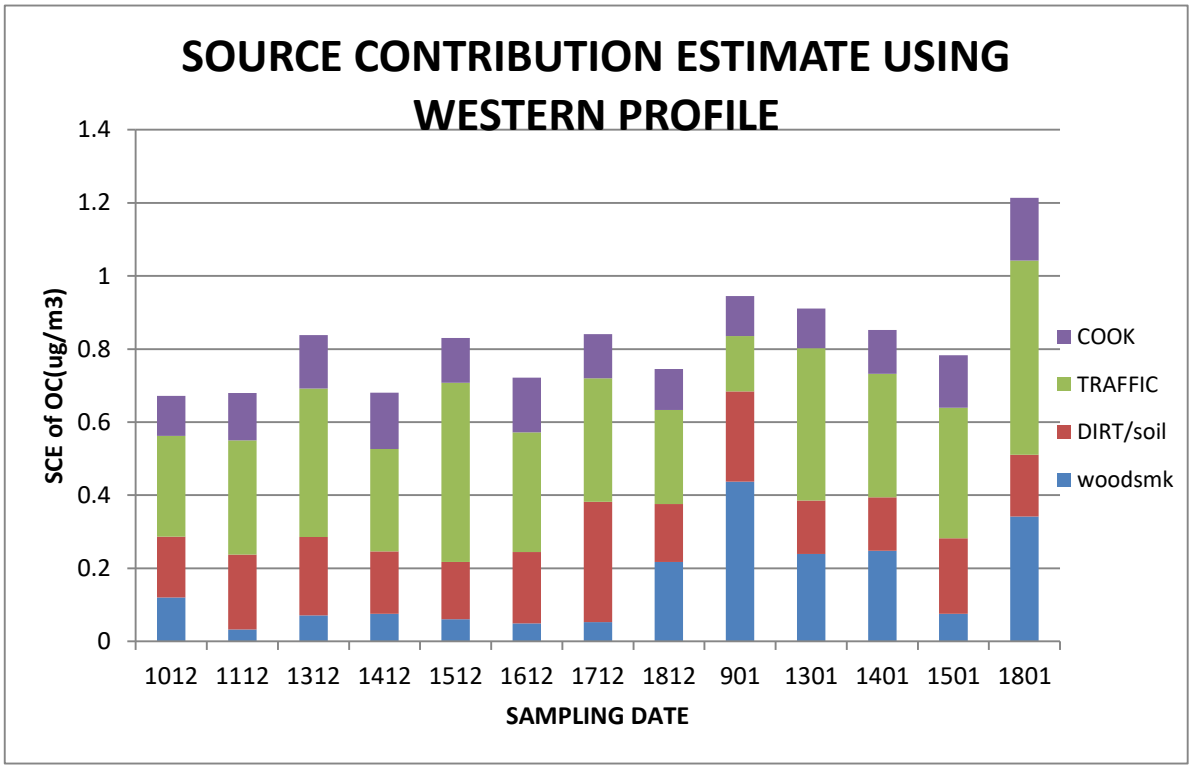


Figure 48 Source contribution using western cooking profile

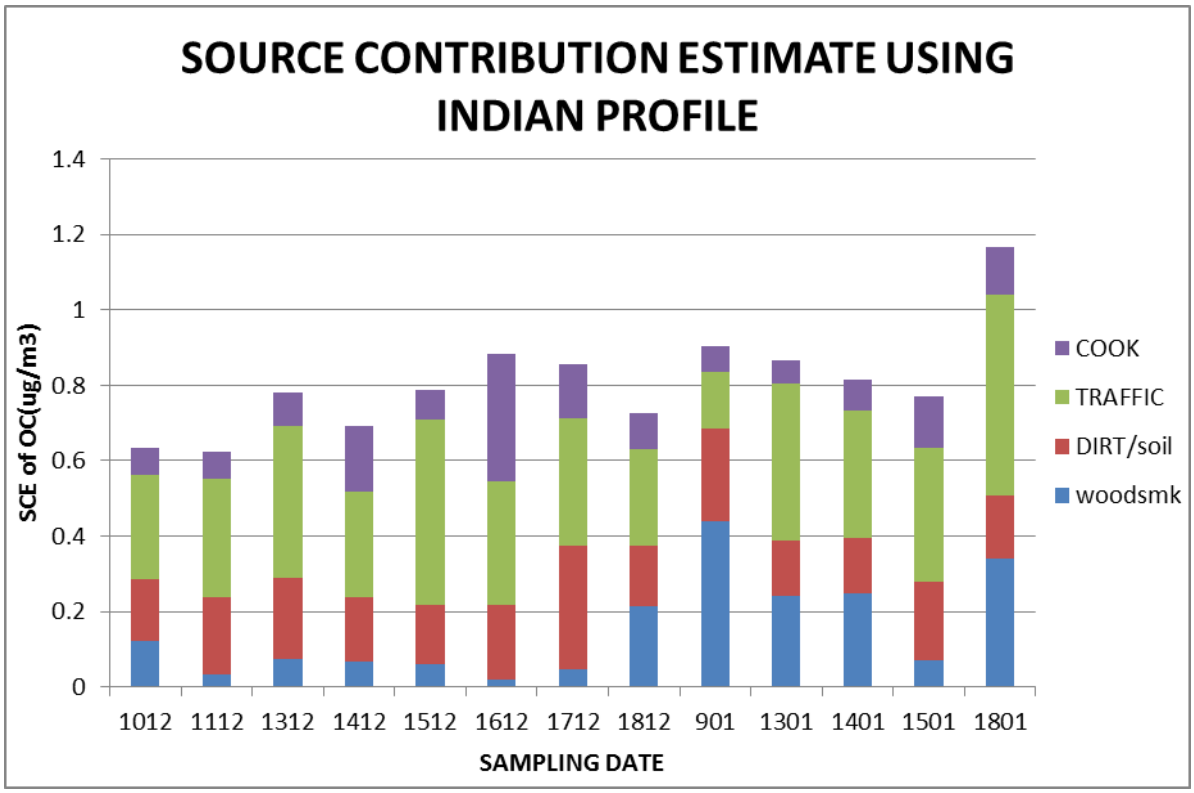


Figure 49 Source contribution using Indian cooking profile

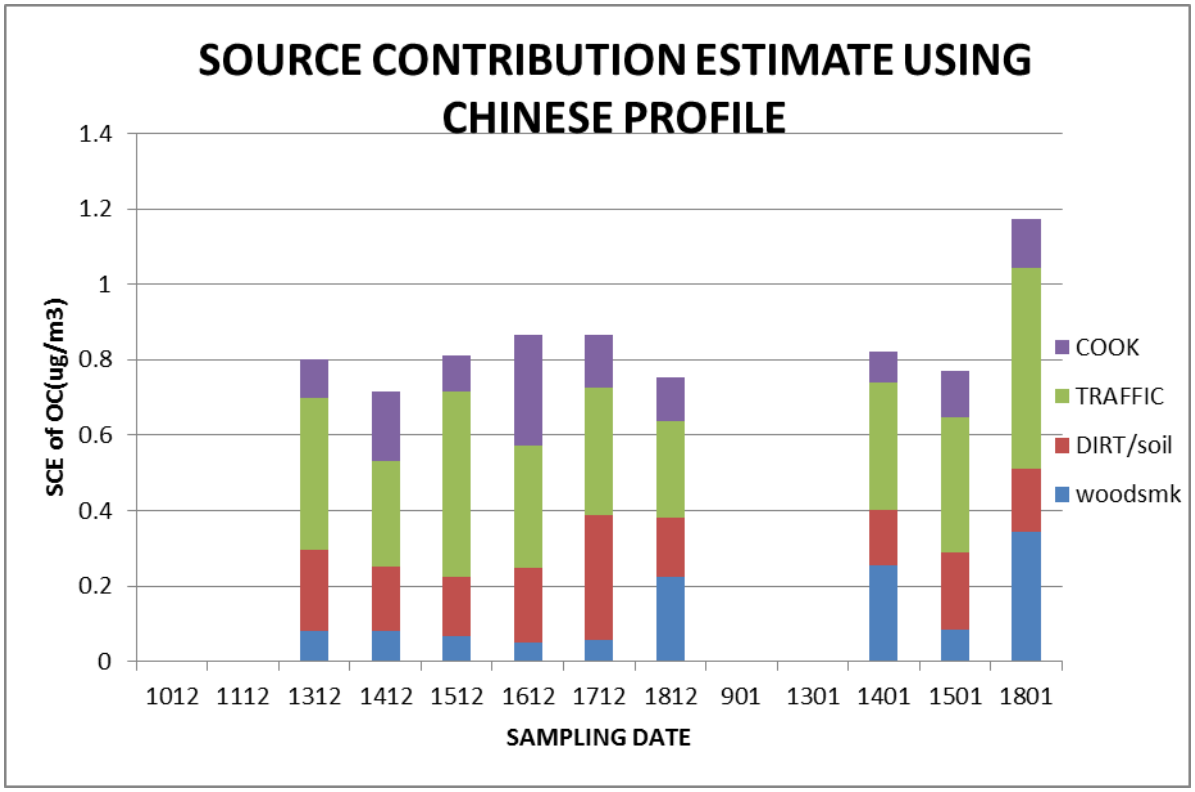


Figure 50 Source contribution using Chinese cooking profile

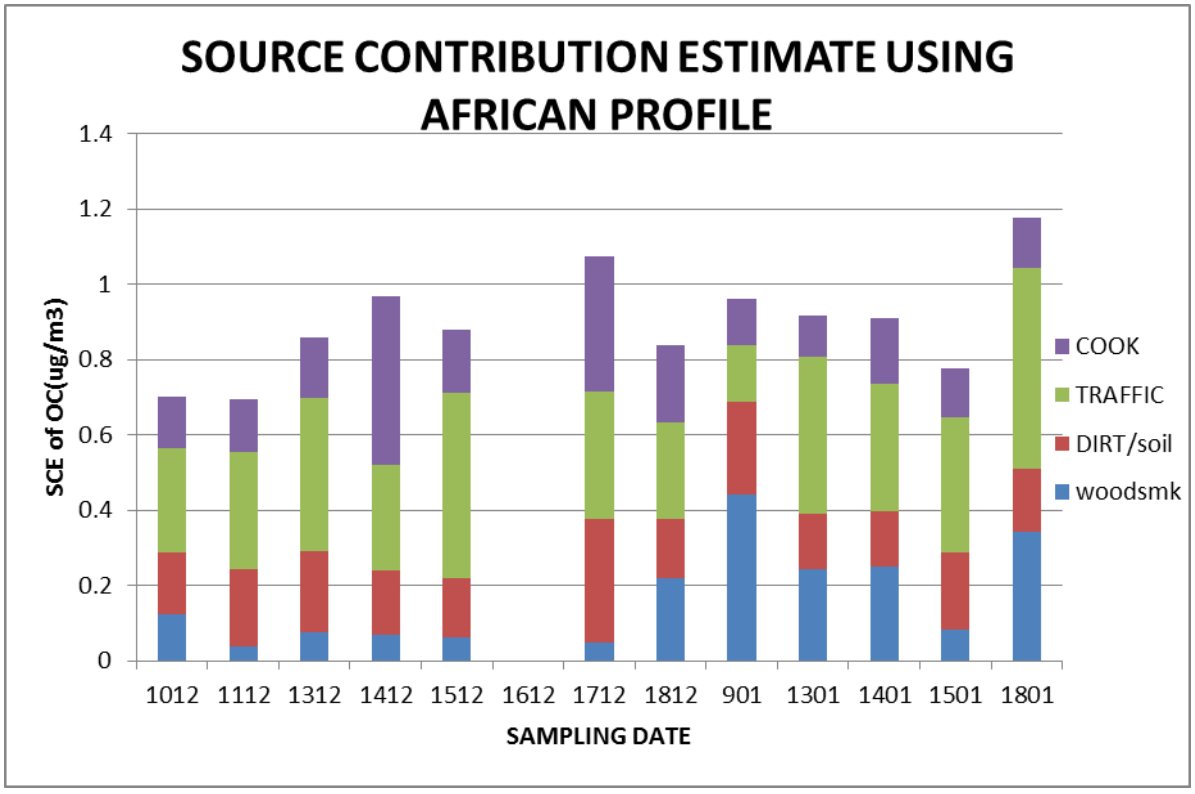


Figure 51 Source contribution using African cooking profile

WEIGHT PERCENT APPORTIONED BY MODEL

Table 51 Percentage mass of organic carbon apportioned by CMB

	% MASS			
DATE	WEST	INDIAN	CHINESE	AFRICAN
1012	42	39.6		43.7
1112	42.5	39		43.3
1312	24.6	22.9	23.5	25.3
1412	48.6	49.5	51.2	69.2
1512	28.6	27.2	27.9	30.3
1612	42.5	51.9	51	
1712	49.4	50.3	51	63.2
1812	53.2	51.8	53.7	59.8
901	78.7	75.4		80
1301	25.3	24		25.5
1401	37	35.4	35.7	39.6
1501	16.7	16.4	16.4	16.5
1801	44.4	44.6	44.8	44.6
average	34.8	34.3	34.9	34.8

Table 52 Daily percent of OC apportioned to source using the various cooking profiles

	AFRICAN PROFILE- percentage				CHINESE PROFILE- percentage				INDIAN PROFILE- percentage				WESTERN PROFILE- percentage			
DATE	woodsmk	DIRT/soil	TRAFFIC	COOK	woodsmk	DIRT/soil	TRAFFIC	COOK	woodsmk	DIRT/soil	TRAFFIC	COOK	woodsmk	DIRT/soil	TRAFFIC	COOK
1012	18	24	39	19					19	26	44	11	18	25	41	16
1112	5	30	45	20					5	33	50	11	5	30	46	19
1312	9	25	47	19	10	27	51	12	9	28	52	11	8	26	48	17
1412	7	18	29	46	11	24	39	26	10	25	40	25	11	25	41	23
1512	7	18	56	19	8	19	61	12	8	20	62	10	7	19	59	15
1612					6	23	38	34	2	22	37	38	7	27	45	21
1712	4	31	32	33	7	38	39	16	5	38	40	17	6	39	40	14
1812	26	19	31	24	30	21	34	15	30	22	36	13	29	21	35	15
901	46	26	16	13					48	27	17	8	46	26	16	12
1301	27	16	45	12					28	17	48	7	26	16	46	12
1401	27	16	37	19	31	18	41	10	30	18	41	10	29	17	40	14
1501	11	27	46	17	11	27	46	16	9	27	46	17	10	26	46	18
1801	29	14	45	11	29	14	45	11	29	15	46	11	28	14	44	14

Table 53 Daily percent apportioned to source using the various cooking profiles (from average concentration)

	Percentage			
	Woodsmoke	DIRT/soil	TRAFFIC	COOK
WEST	17	24	44	16
INDIAN	17	24	45	15
CHINESE	18	24	44	15
AFRICAN	17	24	44	15

5.6 Discussion and analysis of model runs

Four primary pollution sources were apportioned using the average concentration data at Stratford Road that contribute on average 38 % of the particulate organic carbon including traffic, wood smoke burning, food cooking and road dust/soil (model output). The range of total particulate organic matter between the various days was between 17% and 80% as seen in Table 53.

Table 53 represents the percentage of the various contributing sources attributed to the average $PM_{2.5}$ concentration of samples collected during the sampling period, run on the model using the various source profiles (the 4 profiles Indian, Western, Chinese and African). For the average concentration, vehicle exhaust and wood smoke emissions contributed about 45% and 17 % of organic carbon at Stratford Road with food cooking contributing 16% of the organic carbon apportioned. It is observed from this table that across the average, all profiles give similar estimates for estimation of the various sources for instance all attribute 24% to dirt and soil and also 17% to wood smoke.

A closer look at Table 52 shows that on different days there are slight variations in the model outputs on different days when different profiles are used though generally there trends are quite similar. For instance on the 17th of December 2014 cooking was estimated to attribute 33, 16,17 and 14% using the African , Chinese, Indian and Western cooking profile respectively as input in the model run.

The model input included the different range of cooking profile which included Indian, African, Chinese and Western profiles with a general observation of similar apportionment concentrations across the various sources being consistent between runs for the different cooking profiles.

On average a larger contribution of organic carbon was accounted for when the African cooking profile was used, with the Chinese and Indian profile providing similar organic carbon contribution through the sampling period. The source contribution estimates from the various pollution sources were 0.35 µg m^{-3} for traffic, 0.1 µg m^{-3} for food cooking, 0.15 µg m^{-3} for wood smoke and 0.2µgm^{-3} for dust and soil as shown in Table 50. Figure 47 shows other which accounts for other sources of PM which include secondary organic carbon.

Results of the model runs using the various profiles are shown in Figure 48, Figure 49,Figure 50 and Figure 51. From these it is observed that Indian and Chinese profiles apportioned a higher concentration of OC to cooking on the 16th of December, and it is observed that the African profile leads to a general over estimation of apportionment from cooking compared to all other profiles used. Traffic apportionment was found to be highest across all profiles on the 15th of December 2014 and 13th of January 2015. On the 9th of January it was observed that a higher proportion of OC was apportioned to wood smoke as compared to all other days.

On the days there are not data plots the model found colinearity among two or more of the fitting species as such model wouldn't run. This was mainly observed for Chinese and African profiles.

Generally when analyzing all the data and the chi square and r square values in Table 54, the Indian profile show as the best fit for the model runs. The profiles that have best fits are the Indian and Western cooking style profile as observed in Figure 48 to Figure 51. They are also able to run well on all the days as against the African and Chinese profiles. In chapter 3 a good correlation was observed between Indian and Western profiles and it is interesting to observe that these are the profiles that provide consistent results and that give the best fit compared to the other cooking profiles. Generally the amount estimated for the various sources does not vary much among the different profiles but the performance across the various options is clear.

This consistent result could be due to the fact that generally the profiles have similar marker compositions and are found to correlate. Also Stratford road has been shown to be located near mainly Indian and Western restaurants as such the predominant profiles would be those with the most similar composition.

The MPIN matrix was consistent for the different profiles with cholesterol being the species the model used to apportion for cooking Table 49.

On the days of sampling the model has estimated in Table 50 that about 0.16 μg/m^3 of $PM_{2.5}$ was from wood smoke, while 0.15 μg/m^3, 0.55μg/m^3 and 0.1 μg/m^3 were from soil and debris, traffic and cooking respectively. Figure 47 shows the plot of total PM2.5 with the blue section signifying the composition of OC which has not been apportioned by the model and a large amount of it is assumed to consist of secondary organic carbon.

An analysis of previous studies provides insight to the data of the CMB. In Berlin using PMF, similar source contributions to OC were observed during the winter months (6.3–32.2 %) as observed at Stratford Road. Other previous studies had found fine particulate matter ($PM_{2.5}$) comprised of organic matter with concentration of around 25–31 % in the UK West Midlands (Harrison et al., 2004), 21–33 % in Ireland (Yin et al., 2005), 27–47 % in Australia (Chan et al., 1997), 38–47 % in France (Bressi et al., 2013) at sites within and outside Europe, and 50 % in Michigan, USA (Pancras et al., 2013).

Yin et al. analysed two sites in Birmingham in 2010 and identified primary sources found to contribute about 56–85 % on average to fine-particulate organic carbon which included wood smoke, vegetative detritus, natural gas combustion, coal combustion vehicular emissions (diesel engines, gasoline engines, smoking engines), and road dust/soil. The emissions from vehicle exhaust was found to contribute about 57 % of the fine OC, 14 % attributed to other known sources and about 34 % linked to unexplained OC (secondary organic compounds).

They further carried out a study in Southeast England at urban background and rural sites in order to obtain updated and extended information. The samples were analysed with additional markers for food cooking and secondary biogenic aerosols allowing for estimation of the concentration from these additional sources. The CMB model apportioned seven primary sources which explained 53 % and 56 % of the organic carbon (OC) at the urban background and rural sites. The sources apportioned were traffic, wood smoke, food cooking, coal combustion, vegetative detritus, natural gas and dust/soil, when the source tracers for secondary biogenic aerosol was added to the model run a higher proportion of organic carbon was accounted (79 %)(Yin et al., 2015). A good mass closure was observed by Yin et al as they included data for inorganic salts, secondary biogenics and sea salt with 81 % (92 % with the addition of the secondary biogenic source) at the urban background site. Vehicle exhaust was found to be 21 % of the OC and wood smoke 15 %) with food cooking emissions 11 % of OC apportioned. This was similar to the output from the CMB runs using the cooking profiles as observed in Table 53 however the cooking percentage apportioned to cooking was slightly higher (16%) when the new cooking profiles are used.

At a heavily polluted urban site in central California, molecular marker CMB was carried out on ultrafine airborne particulate matter. Meat cooking was identified to account for 33-67% of the $PM_{0.1}$ at the urban site compared to diesel engines which accounted for 15-21%. At a rural site meat cooking contributed 22-26% of the $PM_{0.1}$ OC, and diesel engines accounted for 8-9% (Ham and Kleeman, 2011). As regards the organic carbon of the larger $PM_{1.8}$ particles, meat cooking contributed less to the PM at the rural site than diesel engines; while at the urban site the contribution from meat was still higher than from diesel engines. Lower OC contributions were estimated compared to the measured concentrations, which implies an unidentified contribution of either secondary organic aerosol (SOA) or oxidized primary organic aerosol

(POA). They estimated that meat cooking led to 0.01-0.025 μg/m^3 of $PM_{0.1}$ (Ham and Kleeman, 2011). In the present study 0.1 μg/m^3 of $PM_{2.5}$ is estimated to be from cooking.

In the south-eastern United States, particle phase organic compounds were used in a CMB model and the results indicated that wood smoke, meat cooking and gasoline powered motor vehicles contributed to $PM_{2.5}$ organic carbon concentrations in the range of 25-66%, 5-12% and 0-10% respectively, with minor contributions from paved road dust and vegetative debris (Zheng et al., 2002). Between 2003 and 2004, Zheng et al. (2002) sampled again four sites of the Carbonaceous Aerosol Characterization Experiment (CACHE) and used CMB and carbon isotope analysis to further understand variability of organic components and source contributions to fine organic carbon and $PM_{2.5}$ in the south-eastern United States. Meat cooking was again identified as a primary emission source of OC along with eight other sources including wood combustion (which was the most dominant source, 14-23%), gasoline engine exhaust, diesel engine exhaust, vegetative debris, cigarette smoke, road dust and natural gas exhaust (Zheng et al., 2006).

Meat cooking operations were also identified as one of the sources of ambient fine particulate matter in Houston Texas with a contribution of between 0.9-1.3 μg/m3 at urban sites and 0.7 μg/m^3 at a background sites (Fraser et al., 2003). This is much higher than the 0.1 μg/m^3 obtained from this study but it is good to note that the profile used was from meat cooking and so the profile was quite different to the ones generated (the new profiles were chosen to replicate real cooking with common food choice)

CMB analysis of organic molecular marker data in Pittsburgh Pennsylvania also identified cooking as an anthropogenic source of organic aerosol and $PM_{2.5}$ and found that secondary organic aerosols were actually the major components of organic carbon (OC) in Pittsburgh in

all seasons, whilst primary sources affected ambient concentrations only occasionally (Subramanian et al., 2007).

In Atlanta meat cooking was among the major contributors of fine OC identified with a range of 7-68% (average 36%) in summer periods and 1-14% (average 5%) during the winter months. Gasoline and diesel exhaust contributed 21% and 20% respectively to OC during the summer and 33% to 4% during the winter, with wood combustion being an additional source during that period contributing an average of 50% of OC probably due to use of wood for heating of houses in winter and the festive period.

Lee et al. (2008a) used CMB and UNIMIX receptor models to apportion sources of $PM_{2.5}$ aerosols collected between March 2001 and February 2001 in Korea. The CMB results identified diesel vehicle exhaust as the major contributor to PM (33%), with meat cooking contributing 12% of the $PM_{2.5}$ mass measured. Other sources identified were secondary sulphate (15%), secondary organic carbon (9%), urban dust, Asian dust, biomass burning, sea salt, residual oil combustion, gasoline vehicle exhaust, automobile lead and unknown components (Lee et al., 2008a). The UNIMIX on the other hand only identified seven $PM_{2.5}$ sources and apportioned 30% of the mass to diesel vehicles, 17% to secondary sulphate, 15% from biomass burning, secondary nitrate (13%), gasoline vehicle, secondary organic carbon and Asian dust, but not cooking sources. In Beijing (China), cooking was among the seven emission sources of particulate organic matter identified (Wang et al., 2009). Like other studies, the other sources included gasoline /diesel vehicles and vegetative burning in addition to coal burning in this case. The CMB model established that contribution from cooking was actually higher during the summer, whilst the biomass burning contribution was the highest during the winter (Wang et al., 2009).

An analysis of all the previous studies provide an insight that the profiles generated provide consistent result with food cooking found to contribute about 16% of $PM_{2.5}$ concentrations.

Harrad et al (2003) have used carbon preference indices (CPI) values as useful indicators of the relative contributions to atmospheric concentrations of *n*-alkanes, *n*-alkanoic acids, and *n*-alkanols arising from fossil fuel (e.g. traffic) and biogenic emissions (Harrad et al 2003). For *n*-alkanes, CPI values are expressed as the ratio of the sum of odd carbon number *n*-alkanes to the sum of even carbon number *n*-alkanes. Conversely, for *n*-alkanols and *n*-alkanoic acids, CPI values are the ratio of the sum of even carbon number compounds to the sum of odd carbon number compounds.

The CPI value for alkanes for the sampling period of this study (winter) at Stratford road was 1.44 (C25-C35). Harrrad et al 2003 had CPI values of 1.4 (site a- busy road) and 1.24 (site B-background site) during autumn and winter period (choosen period similar to present study). The CPI values calculated by their study was for C21-C34. The CPI VALUS FOR lkanoic acid obtained from Stratford road was 2.2 (even carbon nmbr compounds to odd carbon number compounds). Previous study by Harrad et al had vaues of 3.07 and 3.16 at sites A and B respectively. The values obtained at this study are similar to figures gotten at site A which is similar site to stratford road site with traffic being a contributing source (petrogenic source) .

Table 54 R2 and chi 2 values for various CMB model runs.

C. Value for daily CMB runs

DATE	1012				1112				1312				1412				1512				1612			
NAME	WEST	INDIAN	CHINESE	AFRICAN	WEST	INDIAN	CHINESE	AFRICAN	WEST	INDIAN	CHINESE	AFRICAN	WEST	INDIAN	CHINESE	AFRICAN	WEST	INDIAN	CHINESE	AFRICAN	WEST	INDIAN	CHINESE	AFRICAN
R SQUARE	0.82	0.82		0.82	0.71	0.71		0.71	0.74	0.73	0.73	0.73	0.78	0.77	0.77	0.77	0.67	0.67	0.67	0.67	0.73	0.72	0.72	
CHI SQUARE	0.07	0.08		0.09	0.08	0.08		0.09	0.1	0.1	0.09	0.11	0.08	0.08	0.07	0.09	0.12	0.12	0.11	0.13	0.09	0.09	0.08	

DATE	1712				1812				901				1301				1401				1501			
NAME	WEST	INDIAN	CHINESE	AFRICAN	WEST	INDIAN	CHINESE	AFRICAN	WEST	INDIAN	CHINESE	AFRICAN	WEST	INDIAN	CHINESE	AFRICAN	WEST	INDIAN	CHINESE	AFRICAN	WEST	INDIAN	CHINESE	AFRICAN
R SQUARE	0.74	0.74	0.74	0.73	0.88	0.88	0.88	0.88	0.94	0.94		0.94	0.84	0.84		0.84	0.86	0.86	0.86	0.86	0.76	0.75	0.75	0.75
CHI SQUARE	0.08	0.08	0.08	0.09	0.08	0.07	0.07	0.08	0.05	0.05		0.05	0.12	0.11		0.12	0.1	0.09	0.1	0.1	0.09	0.09	0.09	0.09

DATE	1801				average			
NAME	WEST	INDIAN	CHINESE	AFRICAN	WEST	INDIAN	CHINESE	AFRICAN
R SQUARE	0.87	0.87	0.87	0.87	0.83	0.83	0.82	0.82
CHI SQUARE	0.09	0.09	0.09	0.09	0.08	0.08	0.08	0.09

D. Average value for all the days run on the model against cooking profile used.

	Average for all the model runs			
COOKING PROFILE NAME	WEST	INDIAN	CHINESE	AFRICAN
R SQUARE	0.8	0.8	0.8	0.8
CHI SQUARE	0.1	0.1	0.1	0.1

5.7 CONCLUSION

Co-linearity between the profiles obtained was found such that each was used in a separate run of the model, rather than attempting to include more than one at a time. The population of the area was found to be culturally diverse, with a substantial community with ethnic origins in the Indian sub-continent. It may also be seen from **Figure 44** that the restaurants in the locality serve a variety of cuisine, with Indian restaurants being the most common.

Concentrations of organic carbon were apportioned in the model, with four primary sources showing a good fit: woodsmoke, dirt/soil, traffic and cooking aerosol. The criterion used for model fitting were the chi-squared and r^2 values, the ratio of the source contribution and standard error (t_{stat}), and the ratio of calculated to measured concentration. The contributions of the four sources according to the cooking style used in the model appear in and show little sensitivity to the input source profile for cooking. There is a large unaccounted mass of OC, labelled in the figure as "other", which we believe is comprised mainly of secondary organic carbon, which is known to make a substantial contribution to OC at UK sites (Harrison and Yin, 2008;Yin et al., 2010; Pio et al., 2011).

Average values of chi -squared and r^2 for the model fits appear in Table 54, and show no significant difference for the compositional profiles tested. Examination of results for individual days showed differences not only between the day-to-day apportionment to sources but also the source contribution estimates obtained when using different source profiles for cooking. However, variations in the model fit as revealed by chi -squared and r^2 values within a day according to source profile were fairly minor Table 54. The day with greatest variation showed a range of r^2 for the different cooking styles of 0.01, whereas the variation between days (of 0.67 to 0.94) was far greater. Similarly there was more day-to-day variation in chi -squared than in the within-day values for cooking styles.

The measured concentration for $PM_{2.5}$ on the days of sampling averaged 6.9 ± 1.6 (s.d.) µg m^{-3} as shown in Table 47. This was a period of unusually clean air for the time of year. The annual mean for the nearest AURN (national network) station of Acocks Green for $PM_{2.5}$ was 12 µg m^{-3} in 2014 and 9 µg m^{-3} in 2015. The mean concentration of organic carbon apportioned to cooking aerosol was 0.12 µg m^{-3} (using the Indian and African cooking source profiles) and 0.13 µg m^{-3} (from the Western and Chinese profiles). This converts to 0.21-0.23 µg m^{-3} organic matter, equivalent to the mass of cooking aerosol particles, contributing 3.0-3.3% of $PM_{2.5}$ mass. This figure compares with a mean mass concentration of OC of 0.39 µg m^{-3}, equivalent to 0.69 µg m^{-3} of cooking aerosol, comprising 4.4% of $PM_{2.5}$ measured at North Kensington, London by Yin et al. (2015) using a CMB model. The Stratford Road, Birmingham samples showed an average contribution from road traffic of 0.37 µg m^{-3} to OC concentrations, equivalent to 0.64 µg m^{-3} (9.3%) of $PM_{2.5}$. This compares with 0.73 µg m^{-3} of OC, equivalent to 1.26 µg m^{-3} (8.0%) of $PM_{2.5}$ at London, North Kensington. These results thus appear very consistent when allowing for the relatively clean air period which was sampled at Stratford Road, Birmingham.

AMS measurements of cooking aerosol have been used by Ots et al. (2016) to estimate a source strength, from which concentrations across the UK have been modelled. Their model predicts a mean concentration of COA in 2012 of 0.5 µg m^{-3} for the model grid cell showing highest concentration (Abdullahi et al, 2017). The annual mean $PM_{2.5}$ at Birmingham, Acocks Green in 2012 was 11 µg m^{-3}. If the cooking aerosol estimated for Stratford Road by CMB is scaled by 11/6.9 to make it equivalent to mean annual conditions for 2012, the concentration is 0.35 µg m^{-3} (taking the mean from all cooking styles). Given the results of comparison of AMS and CMB by Yin et al. (2105) and the possible over-estimation of COA by AMS by a factor of up

to two, discussed in detail by Ots et al. (2016), the scaled concentration of 0.35 µg m^{-3} compares well with the model estimate of 0.5 µg m^{-3} (Abdullahi et al, 2017).

Generally the analysis and the comparison of data obtained from this study with earlier measurements from London (Yin et al., 2015) and with the model results of Ots et al. (2016) show a strong consistency (Abdullahi et al, 2017). This suggests that in recent years in major UK cities, cooking aerosol represents about 3-4% of measured $PM_{2.5}$. The comparison with the numerical model results of Ots et al. (2016) is again suggestive of an over-estimation of COA by the AMS-PMF technique relative to the CMB model results.

The main objective of this section was to compare the estimates of cooking aerosol from the CMB model using source profiles typical of our different cooking styles: Indian, Chinese, Western and African. Despite some differences in the profiles, the CMB model results from each profile are very similar. This may be because in a multi-ethnic cosmopolitan city such as Birmingham no one cooking style is dominant, or because there is sufficient colinearity in the profiles that each leads to a similar estimate, whatever the predominant source of the cooking. The evidence from a survey of local restaurants is that they cater for a very wide range of cuisine, which seems likely to be a dominant factor in this case.

CHAPTER 6 Conclusion Recommendation and future directions.

This thesis presents results of characterisation of PM emissions from cooking with different culinary methods which included Indian, African, Chinese and western style of cooking. During this study rice chicken and potatoes were the major ingredients used as they were found to be the staple food of the large diverse population located in the study area, Birmingham.

Measurements were taken in a controlled environment where no other particulate matter source was present. A kitchen was designed in a trailer located on the University of Birmingham campus where cooking experiments were conducted and particulate matter samples were collected on filters which were placed in the duct of the fume extractor system located above the location where the cooking exercise were taking place. The main food ingredient cooked were similar for all the cooking styles (rice and chicken) with the main difference across the various cooking methods being the methods used to cook as well as ingredients used in the preparation such as spices, oils, peppers.

It was generally observed that the Chinese style of cooking had the highest concentration of PM emissions and this was attributed to the stir frying of ingredients involved in the cooking method. The general order observed for concentration of PM generated from cooking was Chinese, Western, Indian and African and by cooking method, it was found that stir frying generated most particles followed by deep frying and finally stewing (Indian and African cooking was mainly done with a small amount of frying and boiling of the tomatoes used for the Tikka masala and chicken tomatoe stew). The compounds mainly generated from cooking were the glycerides with African cooking emitting the least concentration of these species compared to other culinary methods.

Higher concentrations of heptacosane were observed at the cooking source for Indian cooking with low concentration of tetratriacontane 2.71 µg/m^3 and 0.18 µg/m^3 respectively which was the trend observed for all the cooking types with African generally emitting less concentrations (heptacosane 0.41 µg/m^3 and tetratriacotane 0.07 µg/m^3), highest concentrations were observed in Indian cooking followed by western style cooking then Chinese cooking. A very high concentration of 2.88µg/m^3 was observed for tritriacotane in Chinese cooking, with Indian African and Western style cooking emitting f0.89 µg/m^3,0.4 µg/m^3,1.11 µg/m^3 of the same alkane. This was similar to what was observed in previous studies where the distribution of n-alkanes emitted from Chinese restaurants were generally observed to be substantially different from the distribution from meat cooking (Rogge et al., 1991; Schauer et al., 1999a; He et al., 2004b) and similar to alkane patterns from frying vegetables in seed oils (Schauer et al., 1999a; Schauer et al., 2002). Emission of n-alkanes from cooking consisted of a negligible fraction of the total quantified organic mass emitted and was dependent on the cooking conditions (Rogge et al., 1991; He at al., 2004b). Hildemann et al., (1991a) reported that the n-alkane concentration release rate increased from frying to charbroiling of meat with extra lean meat releasing less compounds than regular meat (Hildemann et al., 1991a). This was similar to observations by Rogge et al., (1991), where charbroiling was found to produce three times the mass of n-alkanes than frying of meat (16 mg/kg of charbroiling meat as against 5.5 mg/kg of frying meat). Rogge et al. (1991) also observed that charbroiling regular meat released four times the mass compared to extra lean meat (thus affected by fat content of meat). In this study the n alkane concentration for Indian and African cooking are found to be a more significant fraction of the total organic mass as seen in Table 25.

Similar to previous studies by Zhao et al in 2007 where Western style fast food cooking had been observed to emit double the concentration of n-alkanes per mg particulate organic matter (POM) compared to Chinese cooking, concentration of alkanes is less in Chinese style cooking

in this study. The n-alkanes have a Cmax at Pentacosane(C25) for western fast food (Zhao et al., 2007a) and meat cooking (Rogge et al., 1991). Chinese cooking exhibits a Cmax at Nonacosane or Hentriacontane (C29 or C31) taken as an indication of the presence of vegetables during cooking operations. In this study Indian cooking had a Cmax at Heptacosane(C27), Western at Hexacosane(C26), African at Nonacosane(C29) and Chinese at Tritriacotane(C33).

Another trend that was observed was that during Chinese and Indian cooking highest concentration of PAH was for dibenz(ah)anthacene 1.96 µg/m^3 and 0.96 µg/m^3respectively. For western cooking highest concentration were found for benzo(b)fluoranthene 1.50 µg/m^3. Generally African food was found to release lower concentrations of PAH than the other cooking styles.

When Chinese cooking and Indian cooking were compared: higher PAH concentrations were observed for Chinese cooking due to stir frying and higher cooking temperature, whilst the Indian cooking style generated the lower PAH concentrations. Indian cooking emitted large amounts of volatile PAH with lower molecular weight like naphthalene, fluoranthene and phenanthrene attributed to low temperature cooking, such as simmering (See et al., 2006). Chinese cooking, on the other hand, was found to emit higher molecular weight PAHs such as benzo[b]fluoranthene, indeno[1,2,3-cd]pyrene and benzo[g,h,i]perylene. These trends were attributed to the cooking methods employed in each type of cooking from the amount of food cooked, the amount and type of oil used, to the temperatures reached during cooking, and cooking time (See et al., 2006).

The effect of the cooking method was also examined by See and Balasubramanian (2008), who found that techniques that involve the use of oil at high temperatures, such as stir frying, pan-frying and deep-frying, released higher amount of PAH compared with those that involve the

use of water, such as boiling and steaming. This is consistent with work of Schauer et al. (2002). Higher quantities of oil are generally used in stir frying, commonly used in Malay and Chinese cooking, than simmering which is the most common technique used for preparation of Indian dishes. In addition, high temperature frying was found to lead to production of higher molecular weight PAHs, while low temperature cooking results in formation of more low molecular weight PAHs (See et al., 2006). McDonald et al. (2003) compared the PAH emissions from charbroiling and grilling meat and found that PAH emissions from charbroiling were about 3–5 times more than when food was grilled. This was attributed to the contact of the lipid material dripping from the meat (during cooking) onto the cooking appliance. Thus, the higher PAH concentrations observed during charbroiling were due to the direct access of lipids onto the hot flame compared to the cooler griddle surface used in grilling (McDonald et al., 2003).

Higher acid concentrations were also observed in Chinese cooking with 9-Octadecenoic acid being the acid with highest concentration for this style of cooking (6.49 $\mu g/m^3$). High concentrations of hexadecanoic acid were also observed in Chinese and all other cooking styles with concentration of 4.22 $\mu g/m^3$, 2.03 µg/m3,1.23 $\mu g/m^3$ and 0.84 $\mu g/m^3$ for Chinese, African, western and Indian cooking respectively. It is important to note that some of the variance observed between the different cooking styles could be attributed to the diffents ingredients used for the diffent cooking styes and themajor method of cooking employed by each cooking method as the study was carried out using basically a single meal type of chicken and rice across all curlinary techniques.

In previous studies by Zhao et al ,Western fast food cooking found that the quantified saturated fatty acids observed a range from C6 to C20 with distinct even to odd carbon preference and a predominance of palmitic acid (Zhao et al., 2007a). Chinese cooking was found to emit C6-C24 fatty acids with a similar even to odd carbon preference and palmitic acid preference similar to meat cooking (Rogge et al., 1991; He et al., 2004b) and seed oil cooking (Schauer et

al., 2002). The most common unsaturated fatty acids observed were oleic acid and linoleic acid for Chinese cooking (Zhao et al., 2007b; He et al., 2004b). The most prominent organic compound released from American cooking is oleic acid (Rogge et al., 1991; Schauer et al., 1999a; Schauer et al., 2002; He et al., 2004b).

The concentration of emitted saturated fatty acids in Western fast food was found to be 13 times higher than in Chinese cooking while unsaturated fatty acid concentrations were only two times higher, attributed to ingredients and cooking temperature. High concentrations of nonanoic acid emissions are observed in both Chinese and Western style fast food cooking with a higher ratio of nonanoic acid to other acids (C8-C10) in Western style fast food. Schauer et al. (1999a; Schauer et al., 2002) compared the emissions of fatty acids from different ingredients, such as meat and vegetables. They found that charbroiling hamburger meat released more fatty acids than frying vegetables. They also found that stir frying released more fatty acids than deep frying.

An analysis of the profiles generated showed that the Indian and western profiles were the most correlated and Chinese and Indian had the weakest correlation, this went on to be observed in the model performance of Indian and Western profiles in the CMB model runs where the best fits were gotten when either of these profiles was used.

The hypothesis that was stated in chapter one that Chinese cooking emits more particulate matter has been clearly proven in chapters 3 and 4 where higher concentrations were observed on periods where Chinese cooking was adopted.

Cooking with the use of gas as the source of heat generally lead to higher concentration of organic compound emitted as was observed in the trailer where both gas and electric were both used. The emissions of compounds emitted at the trailer kitchen(cooking source) using an electric hob was made and the concentrations measured when food was prepared with the

various cooking styles. It was observed that concentrations were higher than when electric hob was used compared to when gas was used for cooking, the average concentration of glycerides cooking Indian food was 0.8µg/m^3 when cooking with gas as against 0.45µg/m^3 when using electric. Acid concentrations also showed a similar trend with average concentration of the group of acids of 0.42µg/m^3 total emitted from gas cooking against 0.2µg/m^3 when electric source of heat was used. For Chinese cooking 1.35µg/$m^3$of total acid as compared to 0.24µg/m^3 emitted when electric was used.

Similar to cooking with gas it was observed that emission during Chinese cooking produced higher concentration of compounds, followed by western style cooking then Indian and finally African. Total concentration for the various cooking methods for glyceride, sterol and acid respectively were found to be; Chinese (0.8, 0.3 and 1µg/m3), western (0.7,0.3 and 0.2µg/m3) Indian (0.5,0.2 and 0.2µg/m3) and African(0.2,0.1 and 0.8µg/m3) showing the trend of total concentration.

Indian cooking had higher concentrations of Alkanes than all other cooking styles with a maximum of 2.71µg/m^3 of heptacosane. For PAH, dibenzo(ah)anthracene was the most prevalent for all four cooking styles. It was found that Chinese cooking led to the release of a higher concentration of PAHs and acids than other styles. The acid with highest concentration for all methods of cooking was hexadecanoic acid.

Particles of mode diameter of particles generated from cooking were found to be largely within the respirable size range, between 15-90nm, with the larger particles generated for cooking methods that involved more water than oil (during stewing). With the use of the AMS it was found that cooking leads to a significant organic loading with grilling as well as stir frying found to have high mass loadings on the instrument. Deep frying was found to lead to a large

number of particles (of smaller diameter than grilling and stir frying) with low AMS mass loading. Cooking with meat (beef or chicken) especially grilling and frying, was observed to lead to higher particles generated than cooking with sea food or vegetables. Water based cooking was found to emit less particles than oil based cooking, with stir frying emitting more PM than deep frying.

An analysis of concentration of PM collected in a real everyday kitchen revealed a similar trend, in terms of generated particle concentration, when meals similar to those cooked in the trailer kitchen was made but the micro environment concentrations were higher than that of the samples collected in the duct of the controlled environment kitchen. This was mainly attributed to the lack of ventilation in the real kitchen. When the concentration of compounds were compared with each other it was found that there was a good correlation for alkane among all cooking methods, African and Chinese had highest correlation for acid compound group and PAH compounds correlations were good but lowest in the combination of African and Indian method. The correlation analysis of the kitchen concentrations with their respective source profile concentrations found a good correlation.

The source profiles were used in the CMB model, 0.2µg/m3 of OC was estimated to be the general concentration of PM from cooking sources at Stratford Road. The profiles that provided the models the best consistently were the Indian and western profiles. This was consistent with what was expected as these were the predominant styles of cooking at the location. The CMB model runs apportioned 16% of the total apportioned Organic Carbon to be from cooking, with traffic, wood smoke and soil debris contributing 44%, 18% and 24% respectively. When the cooking aerosol estimated for Stratford Road by CMB is scaled to make it equivalent to mean annual conditions for 2012, the concentration is 0.35 µg m^{-3} (taking the mean from all cooking styles). Given the results of comparison of AMS and CMB by Yin et al. (2105) and resulting in the scaled concentration of 0.35 µg m^{-3} comparing well with the model estimate of 0.5 µg

m^{-3}. Generally the analysis and the comparison of data obtained from this study with earlier measurements from London (Yin et al., 2015) and with the model results of Ots et al. (2016) show a strong consistency. This suggests that in recent years in major UK cities, cooking aerosol represents about 3-4% of measured $PM_{2.5}$.

With these source profiles generated that represent the geographical location and local emissions, an updated estimation for cooking generated particulate matter contribution to ambient concentrations can be made used as inputs for receptor modelling representing a cost and time-effective alternative for effective control measures.

Policy implications of the research findings include increased monitoring of commercial settings where food is cooked. This can be further effective by the enforcement of fixing exhaust hoods in such locations and ensuring they are used properly. This could go a long way to ensure control of emission from restaurants both indoors and into the ambient air. Also as regulations cannot really be set for individuals to follow in their personal home, with these information, education can be provide to individuals to ensure they are able to be aware of what their cooking choices contribute to their exposure to compounds such as PAHs and they are able to make informed choices on how to limit or control exposure of PM from cooking. Some of the choices that people can adopt are to reduce open grilling and stir frying, boil more food, use extractor fans to direct the particulate out of the houses.

As a next step, the following research is recommended so as to aid in a better understanding of cooking emissions for the establishment of effective control measures to reduce health effects associated and linked with exposure from cooking in homes and cities in the UK and other parts of the world.

- Analysis and identification of the effects of personal exposure to particulate emissions from cooking. Further monitoring of individuals including cooks (over work period and times when they are not cooking) to obtain an idea as to what they are exposed to with analysis of biomarkers.

- The source profile concentrations were emissions measured in the exhaust duct of a capture hood that had the filters removed and so it would be good to obtain the emissions profile in the duct with filters were in place.

- Determining of organic source profile for cooking using other type of ingredients such as beef and sea food.

- Sampling in an area of where there is a wider mix of restaurants and use the data as input for a detailed CMB-based receptor modelling analysis will be undertaken to quantify source contributions using Indian profiles modelling with the source profiles obtained from this study.

- Sampling in kitchens of restaurants where larger quantity food is cooked and where mostly exhaust passed to the ambient air to have a better idea of concentration of emissions from cooking. This will give a better understanding of what cooks and the general public are exposed to as restaurants are all over cities and people do spend a good amount of time in restaurants on days the patronize these establishments.

- Collaboration with the AMS whereby filter samples will be taken concurrently with the instrument and analysis of the filters for organics to determine exact concentrations obtained at same time with the instrument.

- Microenvironment measurements in nearby rooms near kitchens where cooking take place to analyze and characterize the PM generated.

- An analysis of more cooking styles, such as Mexican, Italian among others, would provide further insight to emissions from the different types of restaurants that exist in various cities in the UK.

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